

## APPENDIX L

### SURFACE WATER DISCHARGES

#### INTRODUCTION

Liquid wastes that are generated at FMPC come from three main sources: process water, sanitary sewage, and storm water. Detailed descriptions and diagrams of some of these processes are available (Pennak 1973). These waste streams from the FMPC facility include sump water from the plant production areas, waters from the waste pit area, and waters flowing into the storm sewers from surface runoff over soil contaminated with uranium from spills or deposition of airborne effluents. Liquid effluent streams from FMPC are released to the offsite environment at two locations. These include: (1) The combined sewer and process effluents discharged through the main effluent pipeline at Manhole 175 into the Great Miami River at a point almost directly east of the plant site. This point is about 3 miles (5 km) upstream from New Baltimore; (2) Paddy's Run Creek, a small stream with intermittent flow, lying along the west boundary of the site that joins the Great Miami River approximately 1.5 miles (3 km) south of the FMPC, which received discharges from the storm sewer outfall ditch, and surface runoff from a portion of the production area. The flow in Paddy's Run Creek generally exists only during the period January to May. For the balance of the year it is considered a dry stream bed with occasional flows of a few hours to a day following heavy rains (Patton 1985). Figure L-1 shows the general features of the liquid waste discharge points from the FMPC site.

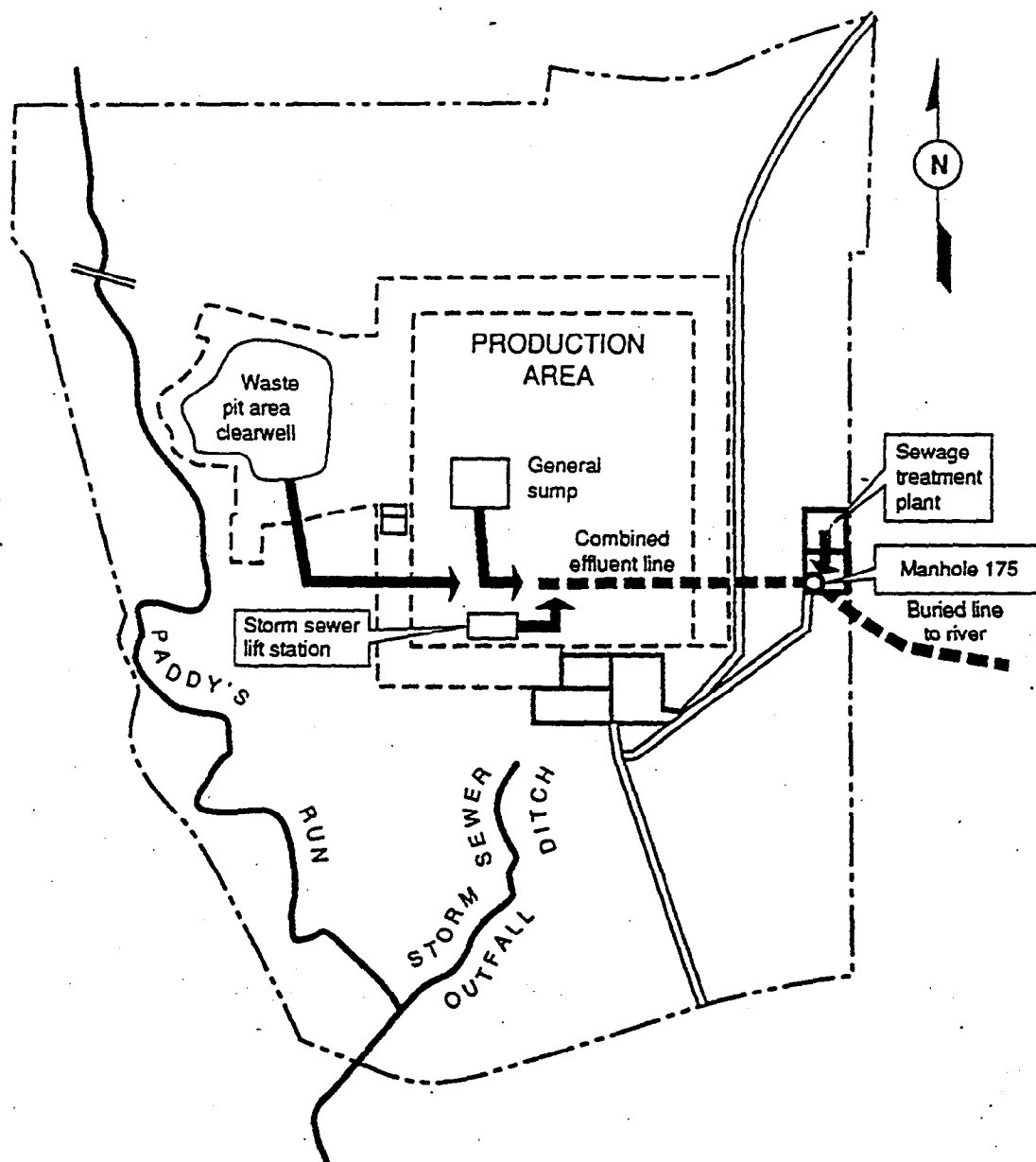
Initially, source term estimates and uncertainties for surface water discharges were derived for the 1960 to 1962 period and presented in an interim draft report (Voillequé et al. 1991). Based on the sources of information and data for that time period, we developed methods for estimating uranium releases to the Great Miami River and to Paddy's Run Creek on a monthly basis. In the present report, we use similar methods of investigation to derive source term estimates for uranium and other radionuclides discharged in liquid effluents from the FMPC for all years of operations. These estimates are reported on an annual basis and the data from original analytical data sheets and other records are tabulated in an annex at the end of this appendix. The tables of daily or monthly data, presented as Tables L1-1 through L1-36 in the annex, will be referenced in the appropriate sections of this report. Much of the background information provided in the interim draft report for the early sixties is presented in this report as well.

#### FACILITIES FOR HANDLING LIQUID EFFLUENTS

##### General Sump System

Each of the individual production plants at the facility had collection sumps and treatment equipment to remove the uranium and thorium from the process waste water. After sampling and analysis was performed to check that uranium content was within pre-

set allowable discard limits (in the sixties, these were  $\text{pH} > 6.3$  and uranium concentration  $< 0.01 \text{ g L}^{-1}$  or  $0.05 \text{ g L}^{-1}$  depending upon the source of effluent) (McCreery 1965), the filtrate was pumped to the General Sump. Thorium wastes were segregated, co-precipitated with barium carbonate and aluminum sulfate to reduce  $^{228}\text{Ra}$  activity and then pumped to the wet chemical pit (Pit 3 until 1968, Pit 5 after late 1968) (Keller 1978). From here the water passed to the chemical waste pit where settling occurred, and the liquid was decanted to the clearwell portion of the pit before discharge through Manhole 175 which carried it by pipe to the Great Miami River.



**Figure L-1.** Liquid effluent flow and discharge points from the FMPC site.

In the early years of facility operation, the General Sump System consisted of three 20,000 gallon receiving tanks (F18-1, F18-2, F18-3), one 5,000 gallon receiving tank (F18-4),

and three 50,000 gallon settling tanks (F18E-1, F18E-2, F18E-3) (NLCO 1957). The settling tanks were installed in late 1956 which accounted for the reduction in contaminants released in the river (Starkey 1958a). The functions of the receiving and settling tanks are summarized below.

#### Three 20,00 Gallon Receiving Tanks:

- F18-1 received effluents from the Refinery sump area, condensate from the digestion area, sampling plant (Plant 1) effluents, and in emergencies, Neutralized Evaporated Product from Plant 2/3.
- F18-2 received Pilot Plant effluents, and when necessary, Plant 8 filtrates.
- F18-3 received waste streams from contaminated sewers of Plants 5, 6 and 9, the Decontamination pad and building, and condensate return to the Water Treatment Plant.

If the uranium concentration was above the limit of  $0.01 \text{ g L}^{-1}$  in these tanks, it was sent back to Receiver Tanks in the Refinery Sump of Plant 2/3 for further processing. If the waste was within the pH and uranium concentration limits, it was pumped to one of three 50,000 settling tanks.

#### One 5,000 gallon Receiving Tank:

Received high fluoride content waste liquors from Plant 4. Then the effluent was either pumped back to the neutralizer tank in the Plant 2/3 Refinery Sump, or pumped to one of the settling tanks.

#### Three 50,000 gallon Settling Tanks:

- F18E-1 and F18E-3 received waste liquid from F18-1, F18-2 and F18-3 where grab samples were taken from the top for uranium analysis. If the uranium concentration was greater than  $0.02 \text{ g L}^{-1}$  it was designated a "rush" sample, and taken to the analytical laboratory for total soluble and insoluble uranium analysis and pH measurements (NLCO 1957).

If the estimated total uranium in the tank was greater than 100 pounds (e.g.  $0.24 \text{ g L}^{-1}$  in 50,000 gallons) it was "mandatory to notify the Plant superintendent" according to the Standard Operating procedures in effect at that time (NLCO 1957). If there were less than 100 pounds of uranium in the tank, the sump supervisor could use his judgment on the possibilities of reclaiming the uranium.

- F18E-2 received Neutralized Evaporator Product (NEP) from Plant 2/3. Samples were taken from a bottom valve. If the concentration was above the limit of  $0.01 \text{ g U L}^{-1}$ , the effluent was sent back to the Plant 2/3 refinery sump. If below the limit, the effluent was pumped to either of the other two 50,000 gallon tanks (F18E-1 or F18E-3).

In 1968, major improvements were made in the General Sump area for waste effluent processing facilities involving the installation of two new 15,000 gallon sludge settling tanks with hopper bottoms and decanting pipes; a new 50,000 gallon sludge settling and decant tank with a flat bottom; and a new head tank for regulated continuous discharge to the river (OHIO 1968).

### Individual Plant Sumps and Normal Operations

The descriptions of the individual plants which follow provide an overview of liquid effluent flow at FMPC. The liquid effluent volume and uranium releases from the various site facilities were provided in monthly loss reports (Yoder 1955, Cuthbert 1960-1961, Marshall 1963, Schwan 1967-1984). Table L-1 provides monthly data on uranium quantities in effluents to the General Sump from the process areas. Although these data are from the early sixties, the relative fraction of uranium discards remained fairly steady over the years.

**Plant 1.** Due to the infrequency of pumping of liquid effluent from Plant 1, effluent was usually pumped to the Plant 2/3 Refinery Sump Receiver Tank (F1-608) for recovery of uranium (Cahalane 1961).

**Plant 2/3.** Three waste streams from Plant 2/3 are important: the sump effluent, the Neutralized Evaporated Product (NEP), and the slag leach slurry from the refinery. While the volume of Neutralized Evaporated Product (NEP) was measured as it was pumped to the General Sump, the Plant 2/3 sump effluent volume was calculated by subtracting the sum of all other individual plant discards into the General Sump from the total volume pumped from the General Sump to the chemical pit. The Plant 2/3 Sump Effluent accounted for roughly 70-80% of the total volume sent to the General Sump, and 25-30% of the uranium in effluents. Table L-1 shows that the NEP waste stream contributed over 60% of the uranium to the General Sump each month, but only 5% of the total volume. The slag leach slurry was pumped directly to the chemical waste pit.

**Plant 4.** Waste liquors from plant 4 which were high in fluorides but rather low in uranium, were pumped directly to the only 5000 gallon tank in the General Sump (F18-4). Routinely, Plant 4 contributed less than half a percent to either the volume or total uranium quantity each month.

**Plant 5.** Liquid waste from the remelt or casting area accounted for approximately 1-2% of the volume, and less than 1% of the uranium, sent to the General sump (Tank F18-3).

**Plant 6.** Contaminated effluents from the machining area were pumped to the General sump (Tank F18-3), contributing on the average 5% of the volume and less than 1% of the uranium to the General Sump. The Heat-Quench Water from the Metal Fabrication Area was pumped directly to the wet chemical pit.

**Plant 8.** Routinely, effluents were pumped directly to the waste pits from Plant 8, and are not listed in Table L-1. In an emergency when discard limits were exceeded, they were pumped through the General Sump (Tank F18-2) for processing and sampling (Cahalane 1961). Because this was an infrequent occurrence, Plant 8 effluents contributed less than a half percent to the volume and uranium totals of the General Sump. However, records summarized in Appendix M indicate that Plant 8 contributed approximately 1200 kg per month directly to the waste pits during 1960, 1961 and 1962.

**Plant 9.** Approximately 1-2% of the volume (< 0.1 % of the uranium) to the General sump (Tank F18-3) contained enriched uranium from Plant 9 (Special Products). The waste stream from the Zirno Slurry was routed directly to the wet chemical waste pit.

**Table L-1. Uranium Discards (kg) to the General Sump From Process Areas<sup>a</sup>**

| Date  | Plant 2/3<br>Effluent | NEP   | Plt 4<br>Cast. | Plt 5<br>Cast. | Plt 6<br>Mach | Pilot<br>Plant | Anal.<br>Lab | Decon<br>Area | Plt 9<br>(Enr) | Total |
|-------|-----------------------|-------|----------------|----------------|---------------|----------------|--------------|---------------|----------------|-------|
| 1960  | 6406                  | 15312 | 35             | 152            | 54            | 920            |              |               | 24             |       |
| 1961  | 5511                  | 17144 | 54             | 81             | 290           | 2830           |              |               | 20             |       |
| 1962  | 3874                  | 4283  | 32             | 108            | 245           | 560            |              |               | 16             |       |
| Total | 15791                 | 36739 | 121            | 340            | 590           | 4310           | 1105         | 85            | 60             | 59140 |
| % of  |                       |       |                |                |               |                |              |               |                |       |
| Total | 27                    | 62    | <0.2           | <0.5           | 1             | 7              | 2            | <0.1          | <0.1           | 100   |

<sup>a</sup> From NLCO 1960-1962.

**Pilot Plant.** Waste effluents from the Pilot Plant refinery, which contained enriched uranium, were pumped to General Sump (Tank F18-2) before being pumped to the pit. Several different waste solutions from at least seven or eight different areas of the Pilot Plant were discharged into the sump including the tin decladding decantation liquors, 3620 area caustic scrub solutions, Winlo filtrate, extraction area raffinate, open air reduction rotocloner scrubber solution, derby shock wastes, and runoff from outside storage pad areas (Cseplo 1961). Only the first two solutions were neutralized to a pH of 7 or higher before being pumped to the sump. Discards from the Pilot Plant were variable from month to month, contributing from as little as 2% up to 10% of the total volume, and from 2% to 9% of the uranium quantity to the General Sump.

Surface and subsurface drainage in the Pilot Plant Area, however, flowed into a manhole on the warehouse storage pad, and then, by gravity, into an open drainage ditch which discharged into Paddy's Run Creek (DeFazio 1962). Analysis of samples indicated that uranium concentrations varied from 7 to 28 ppm with some flows over 5 gallons per minute to the ditch.

**Decontamination Building and Area.** Effluents from this area were variable, but usually contributed less than 1% of the volume, and up to 3% of the total uranium quantity to the General Sump in some months.

**Analytical Laboratory.** Approximately 10% of the volume and 3% of the uranium discharged to the General Sump each month came from the Analytical Laboratory.

There are three process waste streams from the plants which are routed directly to the wet chemical waste pit. They were:

1. Zirno Slurry from Plant 9 (Special products)
2. Heat-Treat Quench Water from Plant 6 (Metal Fabrication)
3. Slag Leach Slurry from Plants 2/3 (Refinery).

### Chemical Waste Pits

Six chemical waste pits have been constructed since operations began at the FMPC. Pits are identified by number based on chronological sequence of their construction, and by type, "dry" or "wet" pits depending upon the main type of material discarded or discharged. Pits 1 (1,080,000 cubic feet) and 2 (351,000 cubic feet) were dry, although some wet materials were added to Pit 2 just prior to completion of Pit 3. Completed in 1959, Pit 3 (6,115,500 cubic feet) was designated a wet chemical pit, and received effluents from the General Sump (Settling Tanks F18E-1, F18E-2, and F18E-3) until it was filled in 1968 (NLCO 1974).

Pit 4 (1,431,000 cubic feet) was built in 1960 as a dry pit. A tabulation of recorded monthly discards of dry and wet wastes to the pits for the time period 1960 to 1962, and annual totals for 1952 to 1974 is located in Appendix M. Characteristics of the waste pits and a description of the methodology used to estimate atmospheric releases from them are given in Appendix K.

In the early years, two overflow lines with valves extended from the "fluoride" pit (Pit 3) to a short tributary of Paddy's Run that lies just west of the pit. In a site review by the US Department of the Interior, Theis (1955) noted that these outlets were apparently not used customarily, and that the tributary and Paddy's Run were usually dry. He did suggest the possibility of groundwater contamination from the waste pits (See Appendix M).

### Sanitary Sewage

The sanitary waste collection and treatment system was a completely separate system from the process waste system. The sewage was treated in a recirculating trickling filter facility, originally sized for 750,000 gallons per day (gpd) but by the late 1970s was receiving only about 125,000 gpd (Keller 1978). The sewage sludge was then incinerated onsite (Pennack 1973). Sampling and analysis were performed on the waste stream before it joined the other effluent streams at Manhole 175. Daily records of waste volume discharged, river flow and calculated concentrations of uranium, nitrates, and fluorides added to the river were maintained, and reported monthly to the Ohio Department of Health (Carr 1955, Walden 1957, Flowers 1960-1961, P&G 1985).

The Chemical Feed Sump from the Water Treatment and the Boiler Plant Area was sampled for Nuclear Materials Control (Starkey 1964a). The results routinely indicated that the stream, although high in volume (approximately 90,000 gallons per day), contributed approximately 5 pounds (2.5 kg) uranium per month to the river.

### Storm Sewer System

The storm water system consists of a grid work of catch basins and about 70,000 feet of buried pipe lines which drains the surface runoff from the immediate vicinity of the processing areas of the facility, a 5,500,000 square foot area (Nelson 1971). Although it was assumed, when operations began in 1952, that the storm sewer system would handle only water, recommendations to install a storm sewer lift station were frequent when sampling of storm sewer drainage indicated uranium contamination. The initial storm sewer system included a storm water detention basin and sump to handle small quantities of contaminated liquids, but no provisions had been made to empty the sump (Quigley 1952).

The detention sump had not been placed in service by February 1954 (Ross et al. 1954). In late 1955, a Storm Sewer Lift Station, located about 2800 feet south and 4100 feet east of the center of the production site (Theis 1955), near the southern end of the system, was installed (OHIO 1955). It was designed to divert and pump waste water flows in the storm sewer system to the process waste discharge line (Manhole 175) to the Great Miami River. A recording flow meter and continuous proportional sampler monitored the discharges, and provided daily data for uranium and liquid effluents discharged to the Miami River from that point (Pennack 1973). Since the storm sewer lift station was not connected to any process, all the uranium lost through it was assumed to be from leaks and spills (Ross 1972). The lift station in place in the early years was designed to take only the initial runoff during a heavy rain. The pumping capacity of the system was approximately 500,000 gallons per day or 350 gallons per minute (DeFazio 1960).

Throughout the late 1950s and 1960s, daily storm sewer samples continued to reflect spills or releases of radioactive process effluents and chemical materials (Starkey 1961a). As a consequence, the majority of the uranium and radioactivity in the combined plant effluent originated from the storm sewer. When the capacity of the storm sewer lift station was reached, water overflowed through the storm sewer outfall to Paddy's Run Creek, a small intermittent stream lying along the west boundary of the site that joins the Great Miami River approximately 3 km south of the FMPC. The volume of storm water that overflowed the storm sewer lift station to Paddy's Run was related to rainfall amounts and patterns. Storm water flow lagged the actual precipitation event by several hours, usually showing an increase in flow the next day (Patton 1985).

Memoranda and various reports suggest growing concern about the liquid effluent handling system at the FMPC from the mid-1950s onward. Table L-2 summarizes the major changes that were proposed and undertaken in response to many of the considerations about unmonitored runoff to the storm sewer and to Paddy's Run. By the late 1960s, water at the Storm Sewer Lift Station was sampled by two proportional automatic samplers: one sampled effluents going to Manhole 175, while the other was activated by an overflow of water going to the storm sewer outfall ditch to Paddy's Run Creek (Nelson 1971). Both samplers were equipped with recording flow meters.

## **DOCUMENTATION OF LIQUID WASTE DISCHARGES FROM FMPC**

Appendix A outlines the sources of information and the types of documents that were found in a variety of repositories around the country for use in the completion of this project. A significant number of documents were related to the liquid effluent system onsite and uranium discharges in liquid wastes from the site because these losses were documented rather thoroughly over the years. Specific documentation is referenced throughout the report. In this section, the documentation used in compiling daily or monthly data for liquid effluent discharges for all years of operation are described briefly.

**Table L-2. Major Changes in the Liquid Effluent Handling System at the FMPC**

| Date      | Modification to System  |
|-----------|---|
| Oct 1951  | First Operations at the FMPC: Storm Sewer System with detention basin and sump installed, but detention basin sump not yet in service in 1954. <ul style="list-style-type: none"> <li>• Process Effluents to River-Measured</li> <li>• All Runoff to Paddy's Run-Periodically Measured</li> </ul> |
| Feb 1954  | Recommendation to install a continuous sampler at the discharge point to the river (MH 175)   |
| Jul 1955  | Storm Sewer Lift Station Installed <ul style="list-style-type: none"> <li>• Process Effluents &amp; Most Runoff-Measured</li> <li>• Some Runoff &amp; Storm Sewer Overflow-Not Measured</li> </ul>  |
| May 1962  | Recommendation to install sampler and flow meter in Paddy's Run near Willey Road at southern plant boundary (Jeffers 1962).   |
| Nov 1965  | Recommendation to install sampler and flow meter at the storm sewer outfall ditch (Starkey 1965c)   |
| Jan 1966  | Installation of pH cell and recorder in Storm Sewer Lift station; alarm sounds in Water Plant when a high or low pH recorded (Riestenberg 1966).  |
| May 1966  | Renovations to outfall pipe to the river so that discharge of the FMPC effluent is in deep portion of the stream (Starkey 1966a).   |
| Aug 1968  | Storm Sewer Ditch Monitor Installed <ul style="list-style-type: none"> <li>• Process Effluents, Runoff &amp; Overflow Measured</li> <li>• Some Runoff to Paddy's Run Not Measured</li> </ul>  |
| Fall 1968 | New tanks installed and key improvements in effluent handling at the General Sump   |
| Jan 1969  | Waste Pit 5 opens, replacing Pit 3 which had been at capacity for months  |
| Apr 1973  | Renovations to outfall sewer to river (CP-73-8) caused by "wear, tear, decay, and action of the elements".  |
| Aug 1986  | Storm Water Retention Basin Installed with capacity of 6 million gallons and emergency spillway overflow at 365 feet.   |

- Original analytical data sheets from the Health and Safety Division for various times from 1954 through 1974 provided uranium, radium and thorium concentrations on a daily, weekly, biweekly or monthly basis on daily or composite samples taken at the MH 175. Similar data sheets provided concentration results for uranium at the Storm Sewer Lift Station.



- "Discharge of Liquid Wastes into the River" (DLW), was a monthly report listing the daily discharge of liquid wastes from the Sanitary Sewer, Storm Sewer, Manhole 175, and Storm Sewer Outfall. Measured volumes and uranium concentrations were listed on a daily basis for these waste streams.
- "Measured Losses and Removals of SS Material From the Production Stream" (MLR) reports, changed to "Routine Operating Losses" report in 1964, provided a monthly summary of uranium discards to the General Sump and stack losses. Volumes and quantities of normal and enriched uranium discarded as liquid waste from each process area are listed for the month. In addition, the MLR reports give the losses to Paddy's Run, discards to the chemical or wet pit, and effluents pumped from the clearwell of the pit to the river. Many of these reports were located covering all years of operations.
- Descriptive reports on key topics were prepared by different departments on a regular basis. Monthly river and effluent flows, and concentrations of uranium and other contaminants in effluents at Manhole 175, the storm sewer, the waste pits, and Paddy's Run outfall were provided in a monthly report, "Comments on Monthly River and Effluent Flow". The Industrial Hygiene and Radiation Department issued monthly reports describing various radiation and air dust studies, stack losses, environmental sampling activities, liquid effluent measurements in the river, and special investigations of problem areas at the facility. Finally, "Aquifer Contamination Control" Reports to the Manager provided quarterly highlights of contamination problems or action taken to improve the effluent control system at the storm sewer, the General Sump, the pit area, the river and the test wells (Starkey 1965a, 1965b, 1967a, 1967b, 1967c, 1968).
- "Comments on Ground Contamination" biweekly reports described ground contamination areas onsite, results of ground contamination surveys of process areas, and charted estimated uranium losses to the storm sewer and rainfall totals for the month. These latter types of reports, which are more descriptive in nature, have been useful in providing background information for conditions that existed at the site in the early years, and in highlighting unusual events and unplanned releases, and are referenced at appropriate locations within the text.

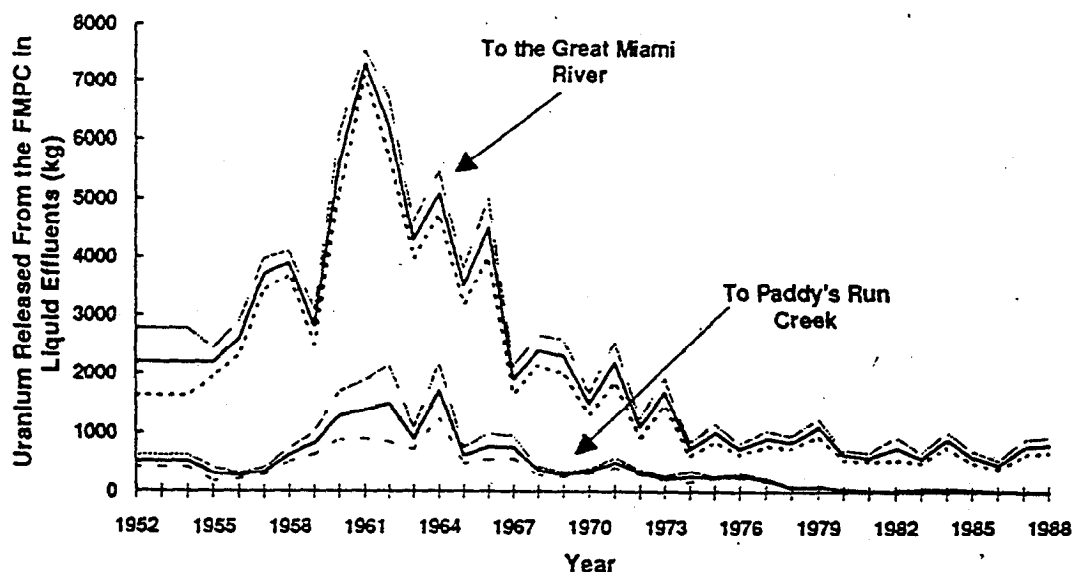
#### **ESTIMATES OF URANIUM DISCHARGED IN LIQUID EFFLUENTS VIA MH 175 TO THE GREAT MIAMI RIVER**

Uranium in liquid effluents leave the FMPC production area by the main effluent line to the Great Miami River or to Paddy's Run Creek via the Storm Sewer Outfall Ditch (SSOD) or runoff from the west side of the production area. Principal contributors to these uranium-bearing effluents included storm sewer runoff, effluent from the clearwell of the liquid waste pit, and treated effluent from the sanitary sewage treatment plant. To calculate the quantity of uranium lost from the FMPC, two key measurements are necessary:

- the concentration of uranium, and
- the volume of effluent to the river (MH 175) or to Paddy's Run.

The total uranium discharged each day via MH 175 to the river was calculated by multiplying the daily uranium concentration ( $\text{mg L}^{-1}$ ) and the volume of water discharged

per day (liters). For Paddy's Run Creek discharges, the measured concentration of uranium and the total volume to the creek taken during specific outfall events, i.e., heavy rainfalls, or for a particular month were used to estimate uranium losses. The uncertainty analyses of these computations are discussed in a later section. Figure L-2 shows the annual uranium release estimates to the Great Miami River and to Paddy's Run Creek for all years. This and the next major sections of this appendix describe the documentation, methodology, and uncertainty analyses computations employed to arrive at these estimates. Data on uranium concentration in liquid effluent taken at MH 175 before discharge to the river are shown in Tables L1-1 to L1-13 in the annex for 1954 through 1969. The results of uranium concentration measurements in the storm sewer and storm sewer outfall ditch to Paddy's Run Creek for 1954 to 1966 are displayed in Tables L1-14 to L1-22 in the annex.



**Figure L-2.** Uranium losses to the Great Miami River via Manhole 175 and to Paddy's Run Creek from the FMPC for all years of operation. The uncertainty of each estimate is described by the 95th percentile (top, broken line), and the 5th percentile (lower, dotted line).

The magnitude of the uranium releases to the river peaked in 1961 with  $7300 \pm 140$  kg uranium. From 1974 onward, the annual releases were below 1000 kg. The uranium losses to Paddy's Run show much more month to month variation than do the uranium loss estimates to Manhole 175. However, the average quantity of 500 kg uranium discharged through Manhole 175 to the Great Miami River each month during the early 1960s (Table L-3) was roughly five times greater than the average quantity of 100 kg of uranium lost to Paddy's Run during that same time (Table L-6). The volume of effluent to Paddy's Run averaged from 2 to 3 million gallons per month during this time period, while Manhole 175 discharged approximately 30 to 40 million gallons each month during the same period (Figure L-3).

Figure L-3 compares the monthly average liquid effluent flow from the FMPC to the river and to Paddy's Run for all years. The average volume of liquid to the river via MH 175 from the FMPC shows a gradual decrease from 30 to 35 million gallons (110 to 130 million

liters) per month in the early sixties to about 15 million gallons (60 million liters) per month in the seventies and eighties. The highest average volume of effluent to the river through the main discharge pipeline (1,400,000 gallons per day) occurred in 1961. Average monthly effluent flow to Paddy's Run is approximately ten times lower than the flow directly to the river, although flow from the site to the storm sewer outfall ditch generally occurs only during heavy rainfall events. The relative difference in flow and variation from month to month can be seen in Tables L1-6 to L1-8, which list the daily and monthly volumes for 1960, 1961 and 1962 to the river, and in Tables L1-18 to L1-22, which list effluent volumes to Paddy's Run for 1960, 1961, 1964 and 1966. These monthly variations in volume are typical of other years as well. Table L1-36 lists the annual effluent volume totals to the river and to Paddy's Run for 1959 to 1984.9

The volume of effluents discharged through Manhole 175 did not show great variation for most months. It was fairly consistent from day to day, showing a gradual decrease over time from greater than a million gallons per day (MGD) in the early sixties to approximately half that volume since 1976.

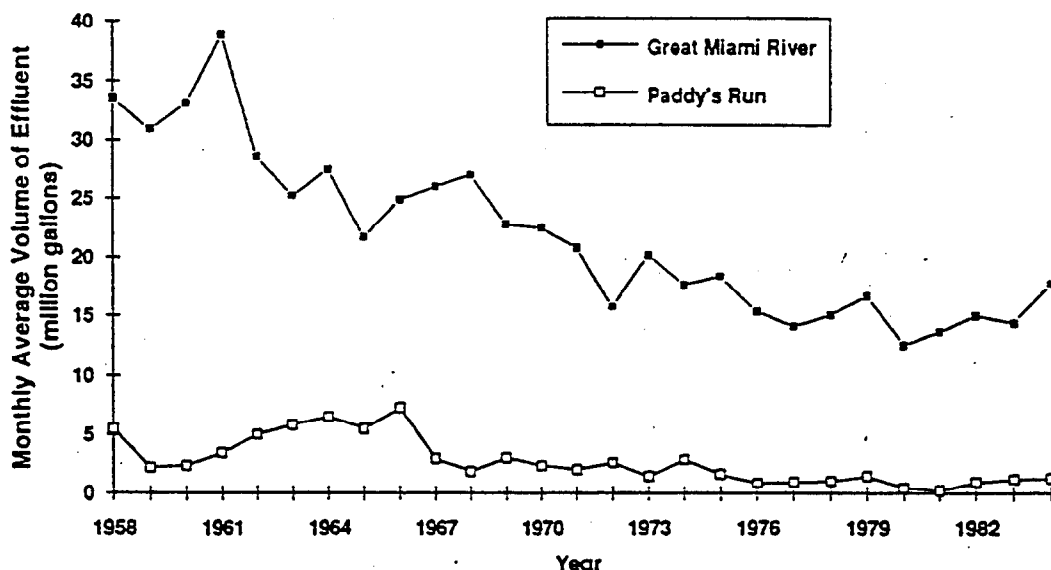


Figure L-3. Comparison of the monthly average volume of effluent to the Great Miami River and to Paddy's Run Creek from 1958 to 1984.

#### Discharges to the Great Miami River Via Manhole 175

Manhole 175 (MH 175), located on the eastern side of the facility, is the discharge point for waste water leaving the site through the main effluent line to the Great Miami River. MH 175 is the final junction point of the major waste effluent streams from the facility. This station is equipped with a recording pH meter, and a Parshall flume flow station equipped with a recirculating sampling line. The discharge flow to the Miami River was continuously measured and a composite sample collected and analyzed on a daily basis. The total uranium discharged each day was calculated by multiplying the daily uranium concentration ( $\text{mg L}^{-1}$ ) and the volume of water discharged per day (liters). The uncertainty analysis of these computations are discussed in a later section.

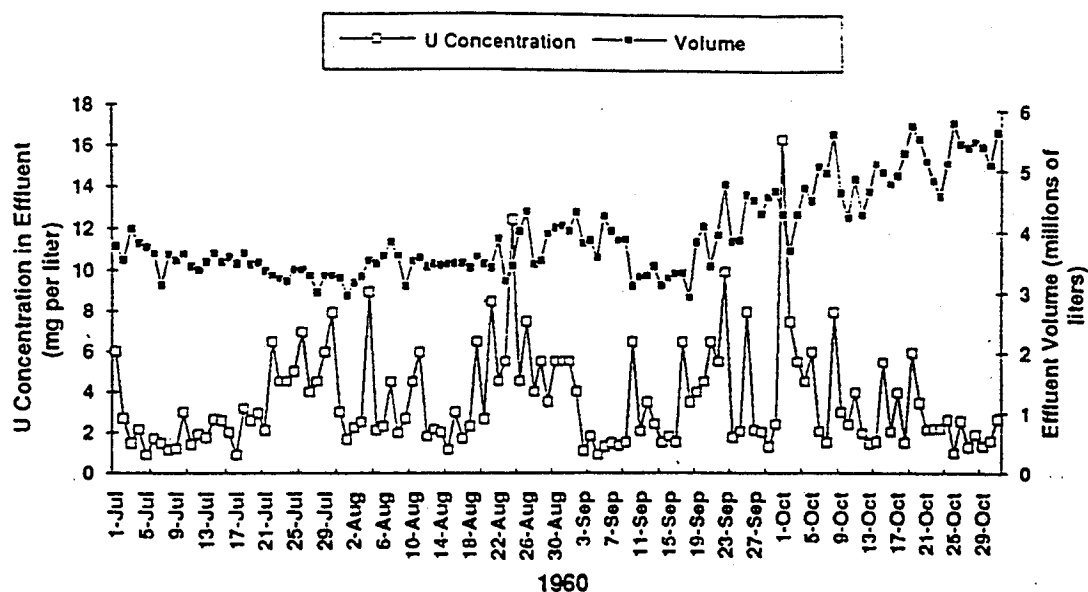
For discharges to the river, both of these quantities were known on a daily or monthly basis for most years of operation, except for 1952 to 1954. Daily uranium concentration measurements on 24-hour composite samples from Manhole 175 for 1954 through 1969 were located, and used in the source term derivation. For the occasional day or month when data sources were not located, an average value for that time period was assumed. Uranium concentration measurements from original analytical data sheets from 1954 through 1969 are listed in Tables L1-1 to L1-13 in the Annex. In addition, Tables L1-6 to L1-8 contain the daily volume measurements from MH 175 to the river. For the interim source term derivation for 1960 to 1962 (Voillequé 1991), daily volume measurements were available for most of 1960 and 1961 (February, April, May, July-December 1960 and January-August 1961) in DLW monthly reports, monthly volume measurements were available from MLS reports (Cuthbert 1960-1961), and from monthly ledger tabulations (Rathgens 1974). An equivalent procedure was followed for all years, with MLS reports, routine operating loss reports and analytical data sheets providing the basis for calculating losses to the river and to Paddy's Run.

Figure L-4 shows the daily uranium concentration and volume measurements taken at MH 175 before discharge to the river for July through October 1960 as an example of the type of variation seen in these parameters. Whereas, daily uranium concentrations varied by a factor of 10 during this period, the effluent volume was more constant. Figure L-5 shows that, over time, the uranium concentration at MH 175 decreased gradually with less variation seen on a day to day basis. The concentration of uranium in the liquid effluent is higher, and shows more daily variation in 1957 than in 1967. In 1967 the daily uranium concentration ranged from 1.5 to 6.6 mg L<sup>-1</sup>, compared to 1957 where concentrations as high as 20 mg L<sup>-1</sup> were seen (See Tables L1-3 and L1-12 in the annex).

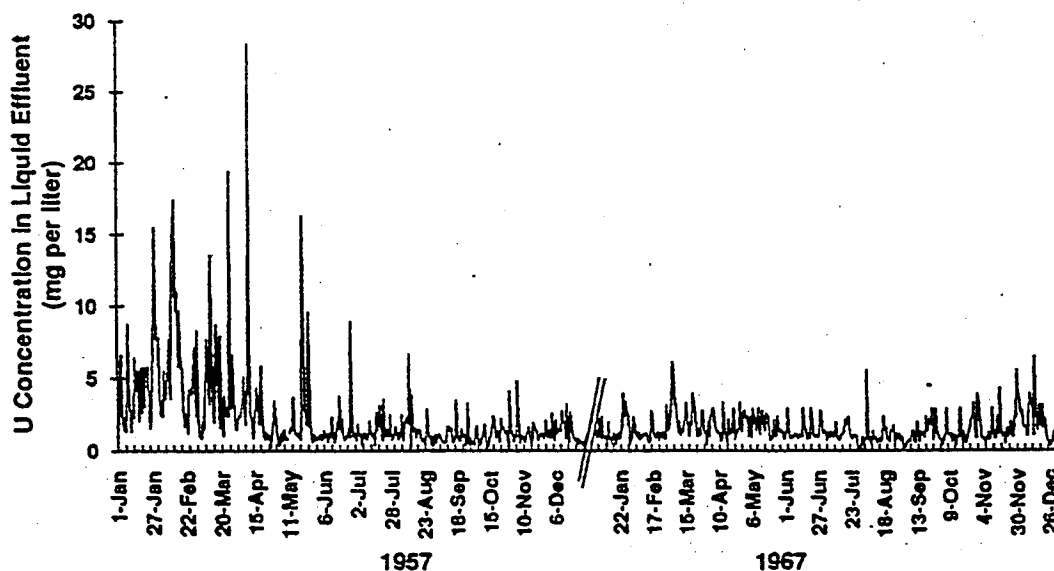
### Uncertainties Associated With Discharges to Manhole 175

Sources of uncertainty for the estimates of losses of uranium through Manhole 175 to the Great Miami River come primarily from the analytical errors in measurement of flow, and in sampling and determination of uranium concentration in the water. Generally, there were differences of 10% or less in the unaccounted-for volume going into Manhole 175 from the various areas onsite. It appeared that the effluent volume to the river was monitored reasonably well (Courtney 1965). Estimates of error for the daily uranium concentration measurements, imprecision in sample preparation for the fluorometric uranium analysis, and volume measurements were made regularly (Brown 1967).

**Uranium Measurements.** For the fluorometric analysis of uranium, the limit of error (LE) at the 95% confidence level was reported as  $\pm 7.1$  mg U L<sup>-1</sup> at the level of 25 mg U L<sup>-1</sup> (28%) in the mid-1960s (NLCO 1966). Control samples indicated the precision and bias of the method for an individual analysis, and were routinely analyzed in a "manner similar to the US AEC GAE program samples". These control samples had a LE of  $\pm 10.3$  mg U L<sup>-1</sup> (bias of +0.2 mg U L<sup>-1</sup>) at the level of 50 mg U L<sup>-1</sup> (21%). The minimum detectable level of uranium by fluorometric analysis was approximately 0.5 mg L<sup>-1</sup>.



**Figure L-4.** Daily uranium concentration (left axis) and volume of liquid effluent (right axis) released to the river for four months in 1960. This figure illustrates the difference in variation seen in uranium concentrations and volume of effluent seen in early years. Whereas the concentration varied by a factor of 10, the effluent volume was more uniform, increasing gradually by a factor of 2 during this period.



**Figure L-5.** Comparison of daily uranium concentrations measurements at the discharge point to the river from 1957 and 1967. The annual average concentration in 1957 was  $2.5 \pm 3.1$  mg L<sup>-1</sup>, compared to that in 1967 of  $1.5 \pm 1.0$  mg L<sup>-1</sup>. The extremes in concentrations decreased in the 1970s and 1980s.

The uranium concentration of 24-hour composite samples from Manhole 175 generally averaged from 2.5 to 5 mg U L<sup>-1</sup>, about 5 times lower than measurements used for LE determinations (Tables L1-1 to L1-13, Annex). Consequently, the relative LE for the Manhole 175 uranium concentration measurements would be expected to be higher as a percentage of the uranium concentration. Based on the measured error limits, and on discussions with individuals from the analytical laboratory at FMPC, the errors associated with the daily uranium concentrations was assumed to be 50% at the 95% confidence level for the 1950s and 1960s. We assume that the daily measurement value represents the mean of a normal distribution of values. Thus the relative standard deviation for each daily measurement is assumed to be 50% divided by 1.96, or 25.5%. For the seventies and 1980s, the relative standard deviation was assumed to be 15%, because of improvements at the MH 175 discharge point and in the analytical procedures.

**Volume measurements.** For flow through Manhole 175, the Limit of Error (LE) for the Parshall Flume flow station was reported as 1.5% of the monthly volume totals in routine quality control reports (NLCO 1966, Brown 1967), although there was no indication whether this was at the 95% confidence limit. Water plant personnel at FMPC generally assumed a variability of about 10% on the daily flow measurements. For these tabulations, a relative standard deviation of 10% on the daily Parshall flume results was assumed to account for measurement error.

For days during a month when daily volume records were not available, the daily average was calculated from the monthly total. The relative standard deviation of daily volume measurements for a month ranged from 6% to 20% for the 18 months in the 1960-1962 period, for which such measurements were available. For those days when an average daily flow was used, a total relative standard deviation of 20% was assumed to account for the normal variation in flow seen throughout the month.

**Total uranium determinations.** The total uranium discharged each day was calculated by multiplying the daily uranium concentration (mg L<sup>-1</sup>) and the volume of water discharged per day (liters). A standard deviation for each daily uranium concentration measurement and volume measurement was calculated by multiplying the daily measurement by the assumed relative standard deviation. The product of the variances of the daily uranium concentration and volume measurements were determined. The standard deviation of the monthly uranium totals was determined using a standard error propagation technique. To determine the 90% confidence intervals (i.e., 5% to 95% predictions) surrounding the estimates, the error was multiplied by 1.645. To illustrate the methodology that was developed previously (Voillequé 1991) to calculate losses to the river for all years, monthly estimates of uranium lost to the river for 1960 to 1962 are shown in Table L-3 with the associated standard deviations. The same method was used to compute the uncertainty of the volume measurements, and those for the 1960-1962 period are shown in Table L-4. Using the same methodology, estimates of uranium released by way of the main discharge point (MH 175) for all years of operations were calculated, and are shown in Figure L-2. The annual estimates are compiled in Table L-5, along with the documentation sources for each year.

For 14 of the 37 years, daily measurements of uranium at the discharge point to the river were used to reconstruct the annual losses of uranium to the river. For other years, except for 1952 and 1953, monthly reports were used. Figure L-6 shows very good agreement for monthly uranium losses to the river calculated from daily analytical data

sheets (ADS), or tabulated from monthly reports for that same period. Hence, the use of monthly reports to provide the uranium loss estimates for our source term reconstruction appears justified by this agreement.

**Table L-3. Monthly Estimates of Uranium Discharged From Manhole 175 to the Great Miami River with Associated Standard Deviations (SD)<sup>a</sup>**

| Month  | 1960   |     | 1961   |     | 1962   |     |
|--------|--------|-----|--------|-----|--------|-----|
|        | U (kg) | SD  | U (kg) | SD  | U (kg) | SD  |
| Jan    | 290    | 20  | 630    | 35  | 480    | 40  |
| Feb    | 340    | 25  | 730    | 40  | 540    | 40  |
| Mar    | 300    | 20  | 730    | 35  | 410    | 30  |
| Apr    | 540    | 40  | 1020   | 55  | 570    | 40  |
| May    | 630    | 40  | 850    | 45  | 480    | 30  |
| Jun    | 530    | 35  | 640    | 35  | 325    | 25  |
| Jul    | 330    | 20  | 530    | 30  | 320    | 25  |
| Aug    | 470    | 30  | 930    | 70  | 380    | 25  |
| Sep    | 380    | 25  | 480    | 30  | 1480   | 240 |
| Oct    | 530    | 35  | 200    | 20  | 390    | 30  |
| Nov    | 540    | 35  | 310    | 25  | 370    | 30  |
| Dec    | 720    | 40  | 300    | 20  | 470    | 50  |
| Annual | 5600   | 300 | 7300   | 140 | 6200   | 300 |

<sup>a</sup> From Voillequé 1991; daily measurements for these monthly totals are compiled in Tables L1-6 to L1-8 in the Annex. These tables illustrate the results of the methodology used to determine uranium quantities discharged in liquid wastes to the river for all years.

**Table L-4. Monthly Estimates of Effluent Volume (million gallons) Through Manhole 175 to the Great Miami River With Associated Standard Deviations (SD)<sup>a</sup>**

| Month  | 1960   |     | 1961   |     | 1962   |     |
|--------|--------|-----|--------|-----|--------|-----|
|        | Volume | SD  | Volume | SD  | Volume | SD  |
| Jan    | 35.2   | 1.2 | 47.0   | 0.9 | 34.2   | 1.2 |
| Feb    | 32.3   | 0.8 | 41.9   | 0.8 | 31.9   | 1.2 |
| Mar    | 31.5   | 1.0 | 45.9   | 0.8 | 31.8   | 1.1 |
| Apr    | 28.8   | 0.5 | 45.1   | 0.8 | 25.2   | 0.9 |
| May    | 30.1   | 0.7 | 42.0   | 0.8 | 24.6   | 0.9 |
| Jun    | 31.1   | 1.1 | 39.0   | 0.7 | 28.5   | 1.0 |
| Jul    | 28.0   | 0.5 | 47.6   | 0.9 | 29.5   | 1.0 |
| Aug    | 29.0   | 0.5 | 46.0   | 1.0 | 31.7   | 1.1 |
| Sep    | 30.3   | 0.6 | 28.1   | 1.0 | 28.4   | 1.1 |
| Oct    | 40.7   | 0.7 | 24.8   | 0.9 | 23.2   | 0.8 |
| Nov    | 38.1   | 0.7 | 28.3   | 1.0 | 23.9   | 0.9 |
| Dec    | 42.2   | 0.8 | 29.9   | 1.1 | 30.1   | 1.1 |
| Annual | 397    | 2.7 | 465    | 3.0 | 343    | 3.6 |

<sup>a</sup> From Voillequé 1991; daily measurements for these monthly totals are compiled in Tables L1-6 to L1-8 in the Annex. These tables illustrate the results of the methodology used to determine the volume of effluent discharged to the river for all years.

**Table L-5. Annual Uranium Losses to the Great Miami River By Way of  
MH 175 With Uncertainty Range (kg)**

| Year | Total U (kg) | 5th %ile | 95th %ile | Information Sources |
|------|--------------|----------|-----------|---------------------|
| 1952 | 2200         | 1600     | 2800      | a                   |
| 1953 | 2200         | 1600     | 2800      | a                   |
| 1954 | 2200         | 1600     | 2800      | a, b, Table L1-1    |
| 1955 | 2200         | 1900     | 2400      | b, Table L1-1       |
| 1956 | 2600         | 2300     | 2900      | b, Table L1-2       |
| 1957 | 3700         | 3400     | 4000      | c, Table L1-3       |
| 1958 | 3900         | 3700     | 4100      | c, Table L1-4       |
| 1959 | 2800         | 2500     | 3100      | c, Table L1-5       |
| 1960 | 5600         | 5100     | 6100      | c, Table L1-6       |
| 1961 | 7300         | 7100     | 7500      | c, Table L1-7       |
| 1962 | 6200         | 5700     | 6700      | c, Table L1-8       |
| 1963 | 4300         | 4000     | 4600      | c, Table L1-9       |
| 1964 | 5100         | 4700     | 5500      | c, Table L1-10      |
| 1965 | 3500         | 3200     | 3800      | d                   |
| 1966 | 4500         | 4000     | 5000      | c, Table L1-11      |
| 1967 | 1890         | 1700     | 2100      | c, Table L1-12      |
| 1968 | 2400         | 2100     | 2700      | d                   |
| 1969 | 2300         | 2000     | 2600      | c, Table L1-13      |
| 1970 | 1500         | 1300     | 1700      | d                   |
| 1971 | 2200         | 1900     | 2500      | d                   |
| 1972 | 1100         | 940      | 1300      | d                   |
| 1973 | 1700         | 1500     | 1900      | d                   |
| 1974 | 720          | 620      | 850       | d                   |
| 1975 | 1010         | 860      | 1200      | d                   |
| 1976 | 730          | 640      | 820       | d                   |
| 1977 | 910          | 780      | 1000      | d                   |
| 1978 | 850          | 740      | 960       | d                   |
| 1979 | 1050         | 960      | 1240      | d                   |
| 1980 | 640          | 560      | 720       | d                   |
| 1981 | 600          | 530      | 670       | d                   |
| 1982 | 750          | 550      | 950       | d                   |
| 1983 | 590          | 510      | 670       | d                   |
| 1984 | 900          | 770      | 1000      | d                   |
| 1985 | 610          | 510      | 710       | d, e                |
| 1986 | 460          | 390      | 550       | d, e                |
| 1987 | 770          | 650      | 890       | d, e                |
| 1988 | 810          | 680      | 940       | d, e                |

a Assume annual totals from 1955.

b Some daily measurements at MH 175 available; NLCO 1954, NLCO 1955, NLCO 1956.

c Based on daily measurements at MH 175, and monthly operating loss reports; NLCO 1957 to 1969.

d From Schwan 1967 to 1983.

e. Annual Environmental Monitoring Reports (Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989).



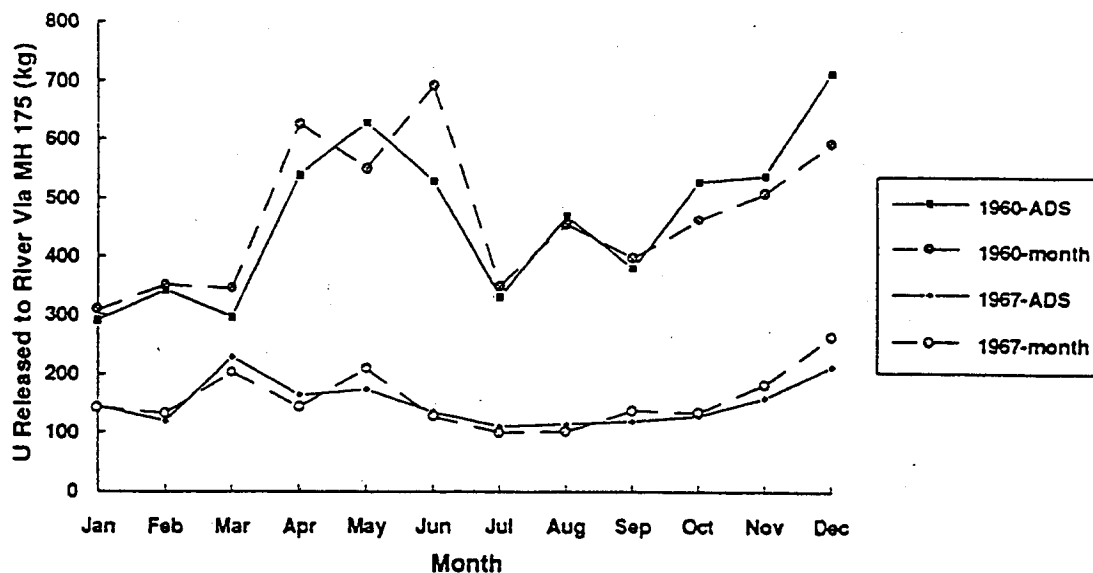


Figure L-6. Comparison of uranium quantities discharged to the river from Manhole 175 for 1960 and 1967, based on daily measurements reported in analytical data sheets from the Bioassay Department (ADS) and from monthly loss reports (month) (Cuthbert 1960-1961, Schwan 1967-1983).

Overall, the quantity of uranium discharged ranged from about 200 kg in October 1961 up to a high of 1480 kg in September 1962. Releases were higher in 1961 than in 1960 or 1962. This is reflected in the annual totals of approximately 5600 kg in 1960, 7300 kg in 1961 and 6200 kg in 1962. These annual totals are 25 to 35% higher than those listed in historic reports from FMPC (Boback et al. 1987). Table L-4 shows the monthly total effluent volumes to the river in 1960, 1961 and 1962. Total flow through MH 175 was higher in 1961, with an average flow rate of 1.3 million gallons per day (MGD), than in either 1960 (average of 1.1 MGD) or 1962 (average of 0.9 MGD).

Uranium releases exceeded 100 kg on at least one day in April 1960 (Table L1-6, annex), August 1961 (Table L1-7, annex), and September and December 1962 (Table L1-8, annex). Losses for the first 9 days of September 1962, which were approximately equal to the total uranium loss for an average month, caused much concern at FMPC (Starkey 1962a). Large releases in 1962 on September 6th (190 kg), 8th (170 kg), and 10th (680 kg), were due to several large accidental releases from Plant 8 during that time. In some months, there was less variation in amounts of uranium discharged per day (for example, December 1960, January 1961), than in other months (for example, September 1960, February 1962). Differences in rainfall patterns and production activities, and the occurrence of spills and unusual releases contribute to the variation. Spills and accidental releases are discussed more thoroughly in an upcoming section.

### Enrichment Categories for Uranium in Liquid Releases

The distribution of uranium among the three uranium enrichment categories changed over time at the FMPC. Of the total uranium released to the river, Figure L-7 shows the fraction of the discharges that were normal, enriched and depleted uranium during each year from 1960 to 1984 (Cuthbert 1960-1962, Schwan 1967-1983). Normal uranium represented the greatest fraction of uranium in the releases until 1967, and from 1970 to 1976. Releases of enriched uranium were minor until 1964 when it reached 40% of the total, and fluctuated between 20% and 60% of the total until 1971. Only a small fraction of depleted uranium was released until 1977 when it rose rapidly to 80% to 90% of the total uranium in liquid effluents. No normal uranium was released after 1978. These relationships of the enrichment categories of uranium in liquid effluents released from the site are quite similar to those for uranium receipts and shipments from the site (See Appendix C).

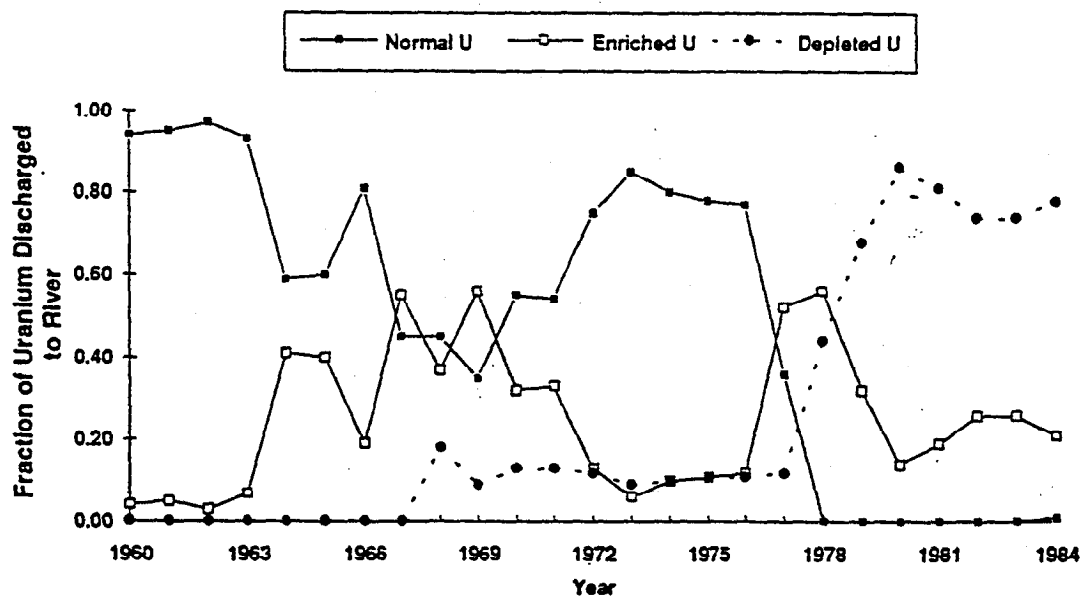


Figure L-7. Relative fraction of normal, enriched and depleted uranium released to the Great Miami River Via Manhole 175 From the FMPC from 1960 to 1984.

### ESTIMATES OF URANIUM DISCHARGED TO PADDY'S RUN FROM THE FMPC

Water collected in the storm sewer system and passed through the storm sewer lift station before being discharged through Manhole 175 to the Great Miami River. A flow meter and continuous sampler monitored the discharges. Since the storm sewer lift station is not connected to any process, all the uranium lost through it was assumed to come from leaks and spills (Ross, 1972). Initially, the storm sewer system had only a detention basin and sump for emptying it when necessary. However, the detention basin was not used, and in July 1955 the storm sewer lift station was installed. Prior to that all runoff from the site went directly to Paddy's Run. The lift station in place in the early sixties was designed to

take only the initial runoff during a heavy rain. The pumping capacity of the pumps was approximately 500,000 gallons per day or 350 gallons per minute (DeFazio 1960).

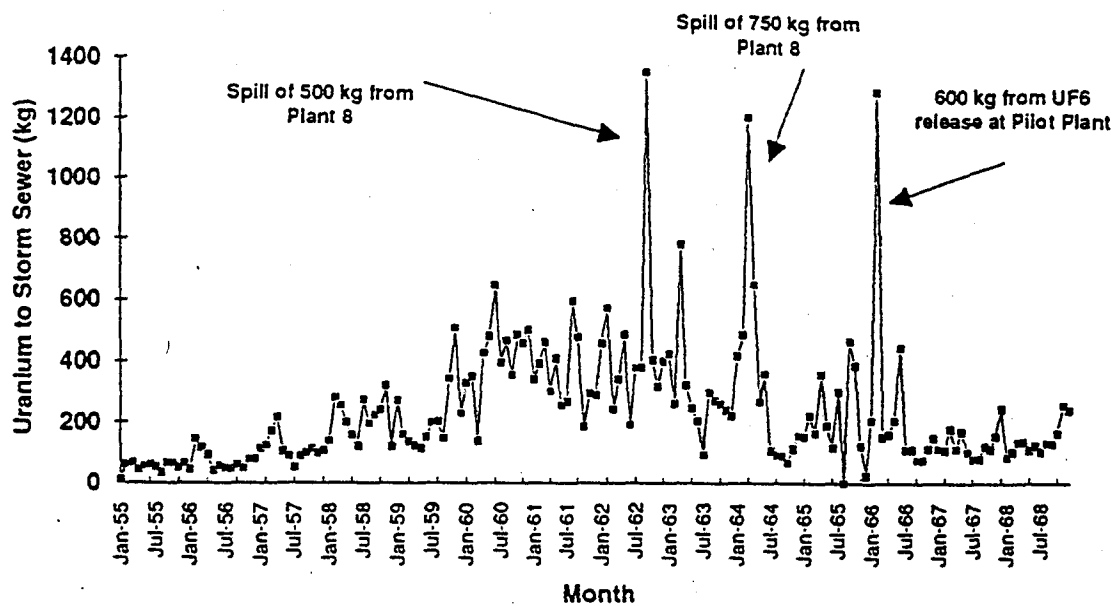
Of the total quantity through the Storm Sewer system, most was discharged through the Lift Station while a percentage overflowed and was discharged through the outfall. Figure L-8 shows the magnitude and variability of the uranium discharges to the storm sewer lift station from 1955 to 1968. The major peaks in September 1962, March 1964 and February 1966 coincide with accidental spills to the storm sewer system, or nonroutine releases of materials (Table L-10). Frequently, uranium concentrations measured at the storm sewer lift station were higher in the late winter or early spring following warmer weather when thawed material in the pipes and on the ground could flow freely. Tables L1-14 to L1-21 in the annex contain the uranium concentrations measured at the storm sewer outfall to Paddy's Run and at the storm sewer lift station from 1954 to 1966. Table L1-23 lists the monthly uranium losses and percentage of total storm water flow that discharged through the outfall and to the lift station for 1960, 1961 and 1962. Clearly, flow to the storm sewer system, and, ultimately to Paddy's Run was quite variable, depending upon total rainfall, and rainfall patterns. Generally, from 2 to over 50% of the flow through the lift station was discharged to Paddy's Run. In some instances, where flow was particularly high, there were reports of up to 80% of the flow being lost to Paddy's Run (Starkey 1964c). Runoff to the storm sewer outfall ditch to Paddy's Run Creek is a major contributor to the uranium contamination in the groundwater to the south of the site. Uranium levels measured in the SSOD and at the lift station are used in Appendix M to develop a source term for groundwater contamination outside of the FMPC.

#### **Estimates of Uranium Losses to Paddy's Run**

Liquid effluent from the site flowed to Paddy's Run when the capacity of the storm sewer lift station was reached. When the capacity of the storm sewer lift station was reached, water overflowed through the storm sewer outfall to Paddy's Run Creek. The volume of storm water that overflowed the storm sewer lift station to Paddy's Run is related to rainfall amounts and patterns. Storm water flow lags the actual precipitation event by several hours, usually showing an increase in flow the next day (Patton, 1985). Furthermore, contaminants were getting into Paddy's Run from areas other than the storm sewer outfall, perhaps from the vicinity of the Pilot Plant storage pad, from the waste pits, or from the vehicle washing station northwest of Plant 1 (Starkey 1959).

Ground contamination occurred on the west side of the Pilot Plant when the sump overflowed the drain to the southwest corner of the site and into Paddy's Run if the rainfall was sufficient (Flowers 1961, Gessiness 1961). By August 1961, curbing had been installed around the sidewalk between the Pilot Plant Annex and the Pilot Plant to direct some of the contaminated runoff to a catch basin, preventing contamination of the soil (Quigley 1961). Pilot Plant personnel made a survey of the ditches and mud holes west of the Pilot Plant, and made note of several large uranium contaminated ditches running to the southwest, eventually discharging into a large gully due west of the Pilot Plant at the second fence (Shaw 1961). In addition, there was a partially excavated hole on the west side of the Pilot Plant which was usually filled with contaminated water. Memoranda indicate that there were plans to pump out the hole (Shaw 1961, Gessiness 1961). It was reported that surface

and subsurface drainage in the Pilot Plant Area flowed into a manhole on the warehouse storage pad, and then, by gravity, into an open drainage ditch which discharged into Paddy's Run Creek (DeFazio 1962). In addition, it was not unusual in the earlier years to drain water from the fluoride pit (Waste Pit 3) directly to Paddy's Run Creek when heavy rains caused high flow in the stream (Starkey 1956).



**Figure L-8.** Monthly quantities of uranium to the storm sewer system from runoff at the FMPC from January 1955 through December 1968. These values were reported in routine operating loss reports from the FMPC. The uranium measured in the storm sewer system comes from leaks, accidental spills and ground contamination events. Nonroutine events involving liquid effluents are recorded in Table L-10.

Prior to the late 1960's, there was no continuous metering of the flow of water through the storm sewer to Paddy's Run Creek (Pennack, 1966), although there was discussion on the continuous measurement of the surface flow in Paddy's Run for some time (Jeffers 1962), and on the purchase of a portable flow meter and sampler (Chapman 1959). In 1966 it was proposed to install a 1,000 gallons per minute (gpm) V-notch weir meter and proportional sampler just downstream from the Storm Sewer Lift Station. Prior to that time, Water Treatment department personnel took grab samples and estimated the flow at the weir notch south of the parking lot (Ross, 1965). Depending upon the duration of the flow, a number of other grab samples would be taken at half hour intervals, and composited. A sample of the composite was then sent to the Bioassay Laboratory for analysis. There continued to be concern regarding the significance of grab samples from the storm sewer outfall in representing uranium quantities lost to Paddy's Run (Quigley 1965). On days when there was a storm sewer outfall flow, the uranium concentration of the outfall sample was usually much higher than the 24-hour composite from the lift station. Analytical results suggested that day-to-day differences in uranium concentrations between the Storm Sewer Outfall grab samples and Storm Sewer Lift Station samples could be significant, but that monthly uranium totals were similar (Ross 1965).

Another source of effluent to Paddy's Run Creek originated as runoff from a portion of the production area near the pilot plant, and as drainage from the waste pit area. In the 1950s, there was a drainage ditch to the south of the waste pits to direct runoff to Paddy's Run (NLCO 1959).

#### Source of Information for Estimates of Uranium to Paddy's Run

For 72 months during the 1960-1966 period, documentation was available that indicated the dates of outfall flows to Paddy's Run, the volume discharged in gallons, and the uranium concentration for each flow to Paddy's Run. Tables L-17 to L-21 in the annex list the losses for those months in 1960-1964 and 1966 where detailed information was located for individual outfall events (Rathgens 1974). The values in the tables come from two types of reports discussed earlier. The first report is "Discharge of Liquid Wastes into the River" (DLW), a monthly report listing the daily discharge of liquid wastes from the Sanitary Sewer, Storm Sewer, Manhole 175, and Storm Sewer Outfall. The Storm Sewer Outfall category lists the dates, volume in gallons, and measured uranium concentration in ppm for each flow to Paddy's Run. For some months, the total number of outfall flows is not known with certainty (e.g., May - Sep 1960), although records of monthly totals of uranium and volume are available for all months (Chapman 1956, Pennack 1973, Rathgens 1974, Bardo 1985, Patton 1985).

The second type of report is the "Measured Losses and Removals of SS Material From the Production Stream" (MLR), a monthly summary of uranium discards to the General Sump and stack losses. Volumes and quantities of normal and enriched uranium discarded as liquid waste from each process area are listed. In addition, the MLR reports give the losses to Paddy's Run, discards to the chemical pit, and "removals" from the pit to the river.

#### Uncertainties of Estimating Uranium Losses to Paddy's Run Creek

The uncertainty associated with estimation of uranium losses to Paddy's Run includes three major components. One area of uncertainty involves unmonitored losses from the site above the point where the storm sewer outfall enters Paddy's Run (where the measured losses were recorded). Records of numerous samples obtained from Paddy's Run indicated that the standards were exceeded in various locations north of where the storm sewer outfall enters Paddy's Run Creek (DeFazio 1960). Quantitative information on the amounts of materials discharged to Paddy's Run from drainage north of the storm sewer outfall location is sparse. One report noted that samples of water in the manhole at the Pilot Plant warehouse showed "uranium contamination but not above what would have been expected normally" (Shaw 1961). The concentration of uranium in the water in the gully was highest at the point due west of Plant 2 and 8 and tapered off at the point west of the Pilot Plant (Shaw 1961). One report noted that the analysis of samples from the open drainage ditch west of the Pilot Plant indicated that uranium concentrations varied from 7 to 28 mg U L<sup>-1</sup> with some flows over 5 gallons per minute (DeFazio 1962).

If these limited data are used to determine whether or not this drainage might be a significant contributor to the total discharges from FMPC to Paddy's Run, then we can calculate the quantity of uranium that would be discharged through this unmonitored

drainage ditch if these conditions existed continuously for a month, and compare that value to our monthly estimates. If we assume that a continuous flow of runoff water of 5 gallons per minute (216000 gallons per month) with an average uranium concentration of 28 mg U L<sup>-1</sup> occurs for an entire month, then we would expect about 20 to 25 kg of uranium per month from this source. This compares to roughly 100 kg of uranium lost to Paddy's Run through the storm sewer outfall ditch each month. Although this rough calculation is conservative, and based on extremely limited data, it represents one source of material loss to Paddy's Run that was not monitored. It may have been the most significant unmonitored source. Consequently, we assume an additional release of 25% above the monthly effluent volume and uranium quantities reported by the FMPC in analytical data sheets and monthly reports.

A second component of uncertainty surrounding the estimation of discharges to Paddy's Run is associated with the collection of grab samples in the storm sewer outfall ditch prior to its convergence with Paddy's Run, and uranium analysis of the grab samples by the fluorometric method. In our interim source term report (Voillequé et al. 1991), data on the number of outfalls to Paddy's Run per month, the volume of water per outfall event, and the uranium concentration of grab samples taken during the overflow event were available for 17 of 36 months in 1960-1962 (See Tables L1-18 and L1-19). Uranium was analyzed by the fluorometric method similar to MH 175 samples. For the individual outfall events in these months, the limit of error (LE) for the uranium concentration measurement at the 95% confidence level was assumed to be 75%, higher than the LE assumed for the uranium determination at the MH 175 discharge point (50%) because the sampling protocol for Paddy's Run involved intermittent grab sampling rather than continuous sampling (Courtney 1965).

Reports indicated that the accuracy of the V-notch Weir flow station ranged from 8% to 15% for normal to flood condition flows, respectively. (Noyes 1966). For this report, the variation is assumed to be 15% for all events. When these errors associated with volume and uranium concentration measurements for individual outfall events are propagated through the month, the LE on the monthly totals range from 4% to 15% of the monthly totals. Consequently, for months when detailed information on number of outfall events was not available, a LE of 15% was assumed for the monthly totals for these 19 months.

A third component of uncertainty for uranium loss to Paddy's Run Creek involves time periods when rainfall, and consequently runoff, were quite high and the capacity of the storm sewer lift station flow meter and V-notch Weir at Paddy's Run may have been exceeded. The water flowing to Paddy's Run occurred when the capacity of the storm sewer lift station was reached. Of the total quantity through the Storm Sewer system, most was discharged through the Lift Station while a percentage overflowed and was discharged through the outfall. Monthly data on measured outfall volume and total uranium to Paddy's Run from the storm sewer overflow indicate that from 2 to 55% of the total flow passed through the outfall to Paddy's Run, with an average of  $21 \pm 11\%$  (Table L1-23).

The pumping capacity at the lift station was approximately 500,000 gallons per day or about 350 gallons per minute (DeFazio 1960). During this time period (1960-1962), there were an average of 3 to 6 times a month when daily flow through the storm sewer lift station was greater than 600,000 gallons per day, with volumes from 750,000 to 850,000 gallons measured occasionally (Starkey 1960-1961). Without specific rainfall patterns and amounts for those specific days, however, it is difficult to speculate whether the flow was

greater than the storm sewer lift station could handle. Based on the occurrence of the storm sewer lift station exceeding its stated capacity roughly 10% to 20% (3-6 times) each month, we assume an additional uncertainty of 20% on the monthly totals of effluent volume and uranium quantity.

These uncertainty estimates for each of the three sources of error that were discussed (unmeasured losses to Paddy's Run, sampling and analytical, and exceeding the capacity of the storm sewer lift station), were incorporated into our final source term estimates for uranium lost to Paddy's Run. Our release estimates, increased by 25% due to unmonitored losses to Paddy's Run, were multiplied by the combined estimates for analytical error and overflow at lift station (15% plus 20%) to provide a bound around each estimate of uranium discharged to Paddy's Run. To determine the 90% confidence intervals surrounding the estimates, the error was multiplied by 1.645. Tables L-6 and L-7 list the monthly quantities of uranium losses and discharge volumes to Paddy's Run for 1960, 1961 and 1962, as an example of the methodology. The uranium concentration data for the storm sewer outfall ditch from original analytical data sheets for 1954 to 1966 are presented in the annex in Tables L1-14 to L1-21.

**Table L-6. Monthly Estimates of Uranium Losses to Paddy's Run With Associated Standard Deviations (SD)**

| Month  | 1960  |        | 1961  |        | 1962  |        |
|--------|-------|--------|-------|--------|-------|--------|
|        | U(kg) | SD(kg) | U(kg) | SD(kg) | U(kg) | SD(kg) |
| Jan    | 160   | 65     | 100   | 40     | 170   | 130    |
| Feb    | 170   | 70     | 100   | 40     | 160   | 130    |
| Mar    | 4     | 2      | 230   | 90     | 390   | 310    |
| Apr    | 40    | 15     | 120   | 50     | 35    | 35     |
| May    | 160   | 60     | 120   | 50     | 160   | 130    |
| June   | 220   | 130    | 80    | 30     | 90    | 75     |
| July   | 170   | 70     | 120   | 45     | 90    | 75     |
| Aug    | 90    | 10     | 20    | 7      | 60    | 45     |
| Sep    | 90    | 30     | 330   | 100    | 6     | 5      |
| Oct    | 110   | 40     | 60    | 90     | 100   | 80     |
| Nov    | 72    | 30     | 140   | 70     | 75    | 60     |
| Dec    | 50    | 20     | 30    | 90     | 135   | 110    |
| Annual | 1300  | 200    | 1400  | 220    | 1500  | 430    |

<sup>a</sup> From Voillequé 1991; measurements for these monthly totals are compiled in Tables L1-18, L1-19 and L1-22 in the Annex. These tables illustrate the results of the methodology used to determine uranium quantities discharged in liquid wastes to Paddy's Run for all years of operations.

For annual losses in the early sixties, the discharges to Paddy's Run were  $1055 \pm 201$  kg in 1960,  $1131 \pm 439$  kg in 1961, and  $1273 \pm 272$  kg in 1962. Few documents listed uranium losses to Paddy's Run routinely, or summarized these losses on a monthly or annual basis. The latest Remedial Investigation / Feasibility Study Groundwater draft report (RIFS 1990), is one of the few documents that lists losses to Paddy's Run. The RIFS report estimates for

losses to Paddy's Run for 1960, 1961 and 1962 are 910, 1180 and 1190 kg, respectively. Our estimates for these years are listed in Table L-8 along with the estimates for all years.

**Table L-7. Monthly Estimates of Effluent Volume to Paddy's Run With Associated Standard Deviations (SD)**

| Month  | 1960                        |      | 1961                        |      | 1962                        |      |
|--------|-----------------------------|------|-----------------------------|------|-----------------------------|------|
|        | Volume<br>(million gallons) | SD   | Volume<br>(million gallons) | SD   | Volume<br>(million gallons) | SD   |
| Jan    | 0.19                        | 0.05 | 3.3                         | 0.5  | 8.9                         | 2.5  |
| Feb    | 9.5                         | 1.6  | 3.4                         | 0.5  | 5.3                         | 1.5  |
| Mar    | 0.05                        | 0.01 | 11                          | 1.5  | 22                          | 6.1  |
| Apr    | 0.64                        | 0.14 | 4.1                         | 0.6  | 1.6                         | 0.44 |
| May    | 0.8                         | 0.04 | 4.1                         | 0.6  | 0.02                        | 0.05 |
| Jun    | 4.9                         | 1.4  | 1.7                         | 0.3  | 1.4                         | 0.4  |
| Jul    | 4.0                         | 0.65 | 3.7                         | 0.5  | 8.4                         | 2.3  |
| Aug    | 0.8                         | 0.15 | 0.35                        | 0.05 | 1.2                         | 0.33 |
| Sep    | 2.9                         | 0.82 | 1.9                         | 0.52 | 0.11                        | 0.03 |
| Oct    | 1.9                         | 0.31 | 0.95                        | 0.26 | 3.3                         | 0.94 |
| Nov    | 1.4                         | 0.22 | 3.6                         | 1.0  | 3.1                         | 0.95 |
| Dec    | 21.5                        | 0.22 | 3.1                         | 0.9  | 4.6                         | 1.3  |
| Annual | 28                          | 2.4  | 42                          | 2.5  | 60                          | 7.4  |

<sup>a</sup> From Voillequé 1991; measurements for these monthly totals are compiled in Tables L1-18, L1-19 and L1-22 in the Annex. These tables illustrate the results of the methodology used to determine the volume of effluent discharged to Paddy's Run for all years.

Figure L-9 compares monthly uranium losses to Paddy's Run from the Storm Sewer Outfall Ditch for three time periods: 1959 to 1962, 1969 and 1970, and 1979 and 1980. The data show that the quantity of uranium lost to Paddy's Run varied considerably from month to month in the early years, so that an average value over a short period of time may not adequately have described a particular month, or several month period. The figure also shows the gradual decrease in total quantity and in monthly variability of uranium released to Paddy's Run. The decline reflects a decrease in production in the seventies and eighties, along with some improvements in the effluent handling system onsite.

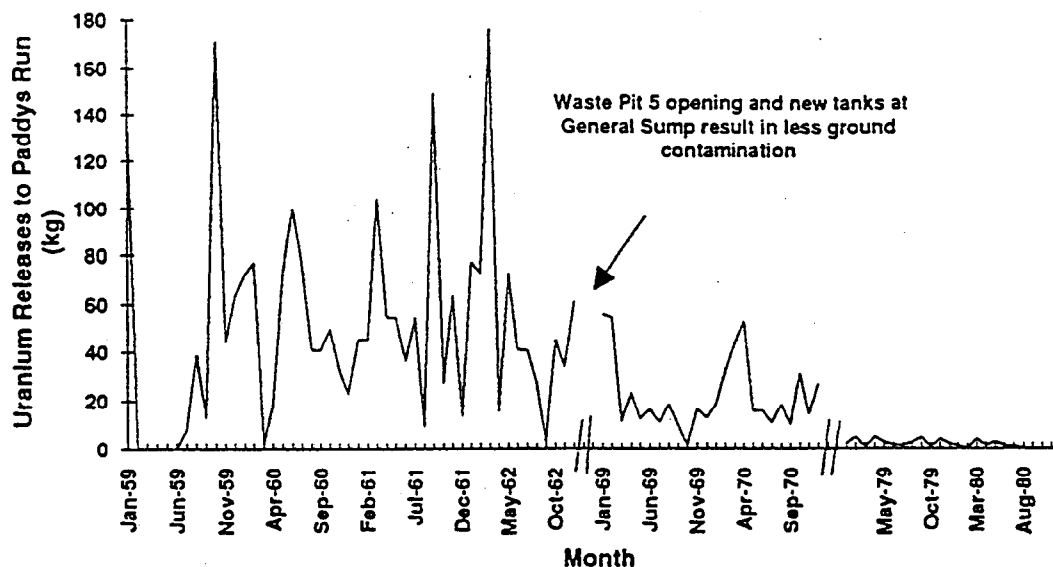
Annual estimates of uranium released to Paddy's Run are shown in Figure L-2 with those releases directly to the river from the FMPC. In Table L-8, estimates of uranium losses to Paddy's Run are listed for all years of operations, with the associated uncertainties.

#### NONROUTINE RELEASES TO SURFACE WATER

Releases of contaminated liquids from spills, drum ruptures, and overflow of sump ponds have been considered in determining the total quantity of uranium released in liquid effluents from FMPC. Regular ground contamination reports were issued on a regular basis. As early as September 1953, an investigation of contamination of the storm sewer outfall to Paddy's Run was conducted after local residents reported changes in the stream from the previous year (Blase and Starkey 1953). The investigators at the site concluded that the primary source of contamination to Paddy's Run was iron salts in runoff from the coal pile.



At that time, all surface drainage from the plant site discharged directly to Paddy's Run via the storm sewer system. During the 1950s, brief "Storm Sewer Contamination" memoranda encouraged plant supervisors to minimize the causes of increased ground contamination and spills (Stewart 1957), but generally no quantitative details of incidents were provided.



**Figure L-9.** Monthly uranium losses to Paddy's Run Creek by way of the Storm Sewer Outfall Ditch for three time periods: 1959-1962, 1969-1970 and 1979-1980. The gradual decline in uranium releases over the years coincides with improvements in the liquid effluent handling system, and with a decline in production activities.

On June 1, 1959, an external area ground contamination survey program of all production plant was initiated on a weekly schedule to inform plant supervision of existing major ground contamination areas, their sources, remedies, and the effect of ground contamination on the storm sewer system (Dodd 1959). Frequently, spills of contaminated materials were described by thickness, and area of gravel covered. For example, a "quarter inch thick" spill covering one square yard, occurred on the graveled area near Plant 4 in February 1964 (Starkey 1964b). Initially all major contaminated areas of soil were to be removed to the waste pits. By 1961, however, the excavation activity was viewed as "not only ridiculous but also an expensive" practice, because of recurring contamination in some locations of the process area (Flowers 1961). With the emphasis on ground contamination, however, the number and extent of spills did appear to decrease over time, shown in Table L-9, in which we have compiled information on the monthly frequency and general source of spills affecting the storm sewer system from 1959 to 1969.

**Table L-8. Annual Uranium Losses to Paddy's Run With Uncertainty Estimates.**

| Year | Uranium(kg) | 5th %ile | 95th %ile | Primary Information Sources |
|------|-------------|----------|-----------|-----------------------------|
| 1952 | 522         | 410      | 630       | a                           |
| 1953 | 522         | 410      | 630       | a                           |
| 1954 | 522         | 410      | 630       | b, d, Table L1-14           |
| 1955 | 300         | 190      | 405       | b, d, Table L1-15           |
| 1956 | 270         | 210      | 320       | b, d, Table L1-16           |
| 1957 | 340         | 280      | 410       | b, d, Table L1-17           |
| 1958 | 630         | 510      | 750       | b, d                        |
| 1959 | 840         | 640      | 1000      | c                           |
| 1960 | 1300        | 800      | 1800      | e, Table L1-18, L1-19       |
| 1961 | 1400        | 1000     | 1600      | e, Table L1-19, L1-20       |
| 1962 | 1500        | 1100     | 2100      | d, Tables L1-17 & L1-18     |
| 1963 | 901         | 720      | 1100      | b, Table L1-18              |
| 1964 | 1722        | 1260     | 2200      | d, e, Tables L1-18 & L1-21  |
| 1965 | 622         | 490      | 760       | b                           |
| 1966 | 771         | 550      | 1000      | d, Table L1-22              |
| 1967 | 753         | 560      | 950       | e                           |
| 1968 | 358         | 280      | 430       | e                           |
| 1969 | 290         | 250      | 340       | e                           |
| 1970 | 349         | 300      | 390       | e                           |
| 1971 | 499         | 410      | 590       | e                           |
| 1972 | 322         | 280      | 370       | e                           |
| 1973 | 231         | 200      | 265       | e                           |
| 1974 | 255         | 210      | 300       | e                           |
| 1975 | 245         | 180      | 250       | b                           |
| 1976 | 272         | 230      | 310       | e                           |
| 1977 | 204         | 170      | 230       | e                           |
| 1978 | 68          | 60       | 80        | e                           |
| 1979 | 84          | 70       | 100       | e                           |
| 1980 | 50          | 40       | 60        | e                           |
| 1981 | 20          | 18       | 22        | f                           |
| 1982 | 20          | 18       | 22        | f                           |
| 1983 | 54          | 40       | 70        | e                           |
| 1984 | 57          | 50       | 70        | e                           |
| 1985 | 39          | 30       | 50        | f                           |
| 1986 | 17          | 15       | 20        | f                           |
| 1987 | <0.5        | <0.5     | 0.5       | f                           |
| 1988 | <0.5        | <0.5     | 0.5       | f                           |

a Assume annual totals from 1954; estimates based on uranium measurements at the storm sewer outfall. the storm sewer lift station not installed until August 1955.

b Based on monthly reports of storm sewer losses; assume 20% to storm sewer outfall ditch.

c Routine monthly reports of operating losses for all months.

d Analytical data sheets for daily losses to storm sewer outfall ditch.

e Monthly records of outfall events to Paddy's Run.

f Annual Environmental Monitoring Reports; assumed uncertainty range of 10%.

**Table L-9. Monthly Frequency and General Location of Spills Affecting the Storm Sewer System During 1959 Through 1969<sup>a</sup>**

| Year | Date   | Number of Incidents   |                                    |
|------|--------|-----------------------|------------------------------------|
|      |        | Affecting Storm Sewer | Areas Involved                     |
| 1959 | June   | 22                    | All processing areas               |
| 1961 | April  | 12                    | All processing plants              |
|      | May    | 14                    | All processing plants              |
|      | June   | 13                    | All processing plants              |
|      | July   | 10                    | All processing plants              |
|      | August | 8                     | All processing plants              |
|      | Sep    | 15                    | All processing plants              |
|      | Oct    | 10                    | Plant 2/3, 6, 8, 9, Pilot          |
| 1962 | Sep    | 16                    | Plant 4, 5, 6, 8, 9, Pilot         |
|      | Nov    | 11                    | Plant 1 pad, 4, 5, 6, 8, 9, Pilot  |
| 1963 | March  | 16                    | All processing plants              |
|      | June   | 7                     | Plant 2/3, Plant 6, Plant 8, Pilot |
| 1964 | Feb    | 18                    | All processing plants              |
| 1965 | Mar    | 4                     | Plant 8, roads                     |
|      | Apr    | 1                     | Railway                            |
|      | May    | 2                     | Plant 2, 4                         |
| 1966 | Jan    | 9                     | Plant 1 pad, 2/3, 8, 9             |
|      | Feb    | 7                     | Plant 2, 8                         |
|      | Mar    | 16                    | Plant 8, tank farm                 |
|      | Apr    | 10                    | Tank farm, Plant 8, 2/3            |
|      | May    | 5                     | Plant 8, 2/3, tank farm            |
|      | June   | 4                     | Plant 2/3, 8                       |
|      | July   | 2                     | Plant 1, roads                     |
|      | Aug    | 4                     | Plant 2/3, Lab Bldg.               |
|      | Sep    | 2                     | Roads                              |
|      | Oct    | 1                     | Bldg. 64 (Th warehouse)            |
|      | Nov    | 2                     | Plant 9                            |
|      | Dec    | 3                     | Plant 8, 2                         |

(continued on next page)

Table L-9. Monthly Frequency and General Location of Spills Affecting the Storm Sewer System During 1959 Through 1969<sup>a</sup> (continued)

|      |      |    |                         |
|------|------|----|-------------------------|
| 1967 | Jan  | 12 | Plant 8, 4              |
|      | Feb  | 11 | Plant 8                 |
|      | Mar  | 3  | Plant 8                 |
|      | Apr  | 4  | Plant 8, 1              |
|      | May  | 3  | Plant 8, tank farm      |
|      | June | 10 | Plant 8, 4, tank farm   |
|      | July | 9  | Plant 8                 |
|      | Aug  | 8  | Plants 8, 2/3, 4, roads |
|      | Sep  | 1  | Plants 8                |
|      | Oct  | 2  | Plant 6, roads          |
|      | Nov  | 4  | Plants 2/3, 4, 8, roads |
|      | Dec  | 4  | Plants 2/3, 4, 8        |
| 1968 | June | 4  | Plants 8, 2, roads      |
|      | July | 4  | Plants 4, Pilot, roads  |
|      | Aug  | 2  | Roads                   |
|      | Sep  | 2  | Plant 8, roads          |
|      | Oct  | 4  | Plants 2, 6, 8, roads   |
|      | Nov  | 1  | General Sump Area       |
|      | Dec  | 2  | Plant 8, roads          |
| 1969 | Jan  | 1  | Plant 8                 |
|      | Feb  | 3  | Plant 8                 |
|      | Mar  | 3  | Plant 8, roads          |
|      | Apr  | 1  | Plant 8                 |
|      | Nov  | 1  | General Sump            |
|      | Dec  | 2  | Plant 8, roads          |

<sup>a</sup> Data were compiled from the monthly reports, "Comments on Ground Contamination in Process Area" (Flowers 1959-1962; Dodd 1958-1959) and "Incidents Affecting the Storm Sewer System" (Riesterberg 1965-1969) that were available for this time period.

From the review of numerous ground contamination reports since 1954, it becomes clear that several locations in the production area continued to be problem areas. These are:

- Plant 8. Contamination prevalent at the east and west end of the plant. Contamination at the north side was caused by the operation of the box furnace. Some of this contamination was checked with the enlargement of the paved area so that it could be flushed from the pavement to the existing sump and storm sewer system (Chapman 1956). Increase in level of storm sewer losses with initiation of the airport scrap handling operation in April 1960.
- Plant 6. The Machining Area from the east pad near the intersection of First and "E" Streets continued to be contaminated from runoff and underground leakage from acid

lines below floor level (Bussert 1956, Tippenhauer 1957). The east pad serves a dual purpose as a plant entrance and a work area, resulting in contamination being spread routinely by vehicles moving through the area (Smith 1961). Although the east pad proper was designed to drain into a sump, "E" street was not so constructed. The lack of curbing on the south end of the pad allowed contamination to drain to the dirt field (Spenceley 1959).

- Plant 2/3. Ore spills common on the SW side. Orange oxide contamination occurs at the SE corner of Plant 2 at the "gulper" station. This problem arose from the muffler discharge connections and from breakage of filter bags in the gulper system (Chapman 1956). Most contamination was restricted to the concrete pad, although the surrounding gravel was replaced after the scrubber system replaced the dry bag collector in late 1956.
- Plant 1 Storage Pad. The area east of the Drum Reconditioning Building usually contained several hundred empty contaminated drums waiting to be baled. Loose contamination fell from the drums onto the pad which flowed into the storm sewer.
- Pilot Plant. The most contaminated areas around the Pilot Plant generally were near the storage pads to the south and west of the Pilot Plant, where the sump overflowed the drain to the SW corner of the facility to Paddy's Run. The small pad near the fence on the west side of the plant was "badly contaminated with piles of  $U_3O_8$ " in the mid-1950s (Chenault 1955). Occasionally, equipment that had been inadequately cleaned was stored on the ground near the SW pad the Pilot Plant (Starkey 1958b). On the west side of the Pilot Plant, the principal contamination was from spills of nitric acid wastes with low uranium concentrations around the nitric acid absorber and storage tank (Davis 1957). In August 1957, a large volume of sump liquor with a low uranium concentration was accidentally spilled while loading the sump truck in that area. This action required "moving a lot of dirt" (Davis 1957). Contaminated soil was removed from near these storage pads periodically, but this area was drained by natural seepage and surface runoff into Paddy's Run Creek.

Over the years, several attempts were made to locate, and thereby eliminate, specific sources of the uranium that were found at the Storm Sewer Lift Station (Chapman 1961, Starkey 1969, Riestenberg 1969, Ross 1972, Lenyk 1977). Generally these surveys indicated that, except for the Boiler Plant area, uranium was entering the storm sewer system plantwide by surface drainage (Lenyk 1977). The main sources of contamination appeared to be the transportation and use of dirty drums, dirty pallets, storage on the ground, and re-drumming operations at some of the storage pads. Furthermore, the use of contaminated oil as dust palliatives on secondary roads and the fly ash pile near the SE corner of the site between the storm sewer outfall ditch and Paddy's Run Creek contributed to storm sewer contamination for years (Karl 1960; Starkey 1960) (See Figure K-1, Appendix K).

For a significant spill into the storm drain, the flow from the lift station could be directed to the General Sump by reversing the flow from the sump, using an emergency gate or diversion valve installed in the early 1970s (Keller 1978). Contamination of this type would usually be washed into the storm sewer system or into Paddy's Run depending upon the location of the contamination. Contamination in Paddy's Run was the primary result of ground spills at the facility (Starkey et al. 1961). The lift station, installed in June 1955,

would handle the majority of the flow in the sewers, with the first fifteen minutes of flow going to the river or catch basins, and the rest flowing over to Paddy's Run (Glass 1955a).

To ascertain the significance of contamination incidents and major unplanned releases of liquid on the determination of the surface water source term, we closely examined reports of incidents involving unusual losses of uranium in liquid effluents, and listed them in Table L-10. The data have been taken from various documents to provide as complete a record as possible of the major accidents or unusual events that discharged quantities of uranium and other radionuclides higher than "normally" released on a daily or monthly basis.

"Notice of Contamination Source" forms were prepared for incidents of chemical spills, radioactive spills, and releases of contaminants directly to the storm sewer due to mechanical problems (Flowers 1960a). The most significant incidents that contributed to possible increases in the uranium quantities in liquid effluent were reported in "Comments on Monthly River and Effluent Flow" reports (Fischhoff 1960-1962). These events were based on the daily calculated uranium losses in the effluent and on formal incident reports received. As the scope of our investigation expanded for all years of FMPC operation, a somewhat similar procedure was followed with the emphasis on those events which may have caused contamination in the storm sewer greater than would be expected from "routine" operations. Table L-10 summarizes the major unplanned releases and losses of material into the liquid effluent system that were reported or recorded in memoranda, daily log sheets, or various types of reports. It provides a brief description of the event, the date, reference source, and general location of the spill or accidental release. The table includes the detailed summary of events for the 1960-1962 period from the Draft Interim Task 2 and 3 report (Voillequé et al. 1991).

The release points for spills or accidental discharges from the FMPC facility would be the same for unplanned as for "routine" liquid effluent releases, that is, through MH 175 to the Great Miami River, or to Paddy's Run. In many cases, the unplanned releases involved quantities of material that were similar in magnitude to daily discharges through MH 175. For example, the incidents on November 21, 1959 (Beers 1960a), January 28, 1960 (Flowers 1960a), and June 1961 (Cuthbert and Quigley 1961) involved the loss of from 2 to 11 kg U, but the main emphasis of these reports was on equipment failure or the need for better procedures.

Occasionally, unplanned releases involved large quantities that were easily measured at the Storm Sewer Lift Station and Manhole 175 (See Figure L-8). For example, in 1962, the uranium concentration measured at Manhole 175 was  $125 \text{ mg L}^{-1}$  on September 10 (about 25 times the concentration measured for routine releases), and  $15 \text{ mg L}^{-1}$  on September 11, reflecting the release of approximately 1000 pounds (450 kg) of uranium to the storm sewer from a digester filter overflow in Plant 8 on September 10. The unplanned releases of September 4 and September 7, 1962 were monitored at Manhole 175 as higher-than-usual concentrations of  $10 \text{ mg L}^{-1}$  on September 5,  $45 \text{ mg L}^{-1}$  on September 6, and  $45 \text{ mg L}^{-1}$  uranium on September 8. This series of losses of materials to the storm sewer system during September 1962 contributed to the highest estimated monthly release of  $1500 \pm 240 \text{ kg}$  ( $\pm$  standard deviation) of uranium via Manhole 175 (Tables L-3 and L-4), compared to the average monthly discharge of about 350 kg in 33 million gallons of effluent.

**Table L-10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC**

| Date<br>(reference)                        | Plant Area                           | Release Amount   | Description of Event or Circumstances   |
|--|--------------------------------------|--|---|
| 9 June 1954<br>(Costa 1955)                | Roadway<br>storage pad<br>to Plant 2 | 871 lb. South<br>African<br>Concentrate  | Transport trailer broke loose from train,<br>spilling contents of 16 drums; cleaned up<br>and drummed.  |
| 6 Dec 1954<br>(Harrell 1954)               | Storage pad                          | Unknown  | Diuranate cake and black oxide in dollies<br>turned over, splitting two drums of<br>diuranate cake  |
| July 1955<br>(NLCO 1955)                   | Plant 1 pad                          | Unknown  | Scrap material spilled over pad due to poor<br>stacking of material and burst drums<br>causing greater contamination than<br>normal of ground and storm sewers.   |
| Oct 1955<br>(Glass 1955a;<br>Stewart 1955) | Plant 2/3                            | Varies from 2 to<br>26 x maximum<br>allowable conc.<br>(MAC) of 0.22<br>dpm mL <sup>-1</sup> . | NW corner of acid recovery contamination<br>by raffinate dumping station to storm<br>sewer; ruptured drums on pad lost to<br>Paddy's Run at the scrap pit.  |
| 1 Nov 1955<br>(Chapman 1955)               | Plant 2/3                            | 26 lb. of U in<br>195,000 gallons  | Loss due to removal and cleaning of vapor<br>lines between denitration and acid<br>recovery   |
| 2 Nov 1955<br>(Glass 1955b)                | Plant 6<br>General<br>sump           | 40 lb. from<br>general sump to<br>river in 20,000<br>gallons.                                  | Refinery sump surge capacity reached so<br>no reprocessing could occur when high<br>levels detected in Tank F18-1. Cause<br>traced to filter problem in Plant 6.  |
| 17 Nov 1955<br>(Chapman 1955)              | General<br>Sump                      | 19 lb.   | Spill of 2000 gallons of calciner feed in<br>Combined Raffinate Area.   |
| 23 Nov 1955<br>(Stewart 1955)              | Plant 2/3                            | 28.9 lb. U in<br>341,000 gallons   | Condensate from denitration vapor line<br>went to general sump prior to analysis;<br>after analysis (10 g L <sup>-1</sup> ), material<br>drummed and returned to refinery.                                |
| 25 January 1956<br>(Strattman 1956)        | K-65 Silo<br>Area                    | Estimated 1000<br>lb. of 2700 lb.<br>insoluble metal<br>oxide that was<br>sent to the silo.    | Metal oxide dust blew out between the top<br>and sidewalk of the first silo, covering<br>several hundred feet around silo; removed<br>with snow layer to concrete trench between<br>Plant 1 and Refinery. |
| 7, 19 Mar 1957<br>(Stewart 1957)           | Storm<br>Sewer                       | 53 lb./day; 10 mg<br>L <sup>-1</sup> at lift station   | Unknown cause; high "U" stream flushed<br>into storm sewer system.  |

(continued on next page)

Table L-10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC (continued)

| Date<br>(reference)            | Plant Area                          | Release Amount  | Description of Event or Circumstances   |
|--------------------------------|-------------------------------------|---|---|
| 3 April 1957<br>(DeFazio 1957) | Roadway                             | Spill material<br>12,000 mg U L <sup>-1</sup>                               | Barrel of material spilled on road at "B" and 2nd Street; material pushed into Storm Sewer manhole.   |
| 22 July 1958<br>(Noyes 1958)   | Drainage<br>system at<br>NE corner  |   | Proposal to modify and repair drainage system surrounding Production Area to eliminate recurrence of flood condition.   |
| 16 Sep 1958<br>(Ross 1958)     | Refinery<br>Area                    | 8.32 mg L <sup>-1</sup>   | Spill of raffinate in refinery area showed a U concentration of 4100 mg L <sup>-1</sup> ; rain washed spill to storm sewer and Paddy's Run.   |
| 23 July 1959<br>(Harr 1959)    | Plant 2/3                           | 1000 lb. U; about<br>400 lb. to storm<br>sewer                              | Release of hot uranyl nitrate solution from the 8" vent of the #212 sparge tank on to the denitration pad, the roadway east of the Refinery and the gravel area east to Plant 4. Gravel excavated to pit. |
| 21 Nov 1959<br>(Beers 1960a)   | Plant 8<br>Storm<br>Sewer           | 500-750 gallons<br>of 1800 mg L <sup>-1</sup><br>U; 12 lb.                  | Digestion filter pump failure   |
| 5 Jan 1960<br>(Flowers 1960a)  | Source<br>unknown                   | 46 kg (101 lb.)   | Detected in storm sewer and MH 175 samples; concentration 12 mg U L <sup>-1</sup> .   |
| 28 Jan 1960<br>(Flowers 1960a) | Plant 8                             | 11 kg (24 lb.)  | Not given   |
| 18 Feb 1960<br>(Flowers 1960b) | Plant 8 to<br>Pit 3                 | "Unknown"<br>(MAC not<br>exceeded in<br>Paddy's Run)                        | Effluent line from Plant 8 broke near entry to Pit 3; flow to Paddy's Run via drainage ditches  |
| 29 Aug 1960<br>(Harr 1960)     | General<br>Sump                     | 111 lb. U to<br>waste pit   | One of tanks (F18E-3) was pumped too pit before analysis.   |
| 1 Oct 1960 (Beers<br>1960b)    | Plant 8<br>Storm<br>Sewer           | 70 kg (155 lb.<br>UO <sub>3</sub> )   | Not clear; 16.5 mg U L <sup>-1</sup> detected in storm sewer and MH 175 samples.  |
| 20 Feb 1961<br>(Starkey 1961a) | Pilot Plant,<br>west side           | Not given in<br>report  | Process and contaminated water pumped onto ground; area "cleaned up".   |
| 20 Mar 1961<br>(Bravard 1961a) | Sump Area,<br>Plant 9 "D"<br>Street | Spill material<br>had 1 g U L <sup>-1</sup> ; "2<br>-3 mR h <sup>-1</sup> " | Overflow of sump pit that empties filtrate hold tank diked area to graveled area covering 10' by 40'.   |

(continued on next page)



**Table L-10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC (continued)**

| Date<br>(reference)                                    | Plant Area                   | Release Amount   | Description of Event or Circumstances  |
|--|------------------------------|--|--|
| 27 Mar 1961<br>(Bravard 1961b)                         | Plant 9, So.<br>gravel area  | 150 kg (330 lb.);<br>10 gal drum<br>black oxide                    | 55-gal drum with 10-gal drum inside<br>failed when burning briquettes added; area<br>cleaned up.   |
| 28 Apr 1961<br>(Beers 1961)                            | Plant 8 UAP<br>Acid Filtrate | 158 kg (347 lb.)<br>U; 830 gal of 50 g<br>U L <sup>-1</sup> .      | Spill contaminated 40-50 yards of gravel;<br>storm sewer was closed and material was<br>drummed  |
| Jun 1961<br>(Cuthbert &<br>Quigley 1961)               | Pilot Plant,<br>outside      | 1.5 kg (3 lb.) U   | Area SW of Pilot Plant; material removed<br>to waste pit.  |
| 4 Sep 1962<br>(Gessiness 1962)                         | Plant 1<br>storage pad       | 91 SS kg U (200<br>SS lb.)   | Leakage from drums of contaminated<br>solvent being transported to digestion.  |
| 7 Sep 1962<br>(Gessiness 1962)                         | Plant 1<br>Storage Pad       | 307 SS kg (675<br>SS lb.)  | Leakage from drums of contaminated<br>solvent being transported to digestion.  |
| 10 Sep 1962<br>(Noyes 1962a;<br>Strattman '62;)        | Plant 8<br>Storm<br>Sewer    | 455 kg (1000 lb.)<br>U in 1820 gallons                             | Winlo digestion filter overflow of liquid<br>containing UO <sub>2</sub> Cl <sub>2</sub> .  |
| 13 Dec 1962<br>(Beers 1962;<br>Noyes 1962b &<br>1962c) | Plant 8<br>Storm<br>Sewer    | 70 SS kg (153 lb.)<br>enrich U, 92 SS<br>kg (203 lb.)<br>normal U. | Calculated release based on storm sewer<br>sample from MH 23 and digester sample in<br>Plant 8; due to plugged filtrate line to<br>precipitator. |
| 1-10 Mar 1964<br>(Starkey 1964b)                       |                              | 1640 lb. U to<br>Paddy's Run                                       | Not clear; probably involved Plant 8.  |
| 14 Feb 1966<br>(Starkey 1966a)                         | Pilot Plant                  | 1230 lb. U   | UF <sub>6</sub> release.   |
| 6 Jun 1966<br>(Nelson 1966)                            | Plant 2                      | 900 lb.  | Process "slop" liquor leaked from diked<br>area beneath the NE and SE hold tanks on<br>N side of refinery.                                       |
| 2 Aug 1966<br>(Noyes 1966)                             | Plant 2                      | 600 lb. of U at<br>1.12% <sup>235</sup> U                          | Open nitric acid valve to NE hold tank<br>allowed overflow of materials with U<br>concentration of 50-70 g L <sup>-1</sup> to storm<br>sewer.    |
| 12 Oct 1966<br>(Starkey 1966b)                         | Plant 3                      | 100 lb. U onto<br>graveled area                                    | Leaking overhead line near the SE corner<br>of the plant; some gravel was removed for<br>reprocessing.   |

(continued on next page)

**Table L-10. Major Unplanned Liquid Releases and Spills to the Onsite Liquid Effluent System at the FMPC (continued)**

| Date<br>(reference)                            | Plant Area               | Release Amount  | Description of Event or Circumstances  |
|--|--------------------------|---|--|
| January 1967<br>(Riestenberg 1967)             | Plant 8                  | Various   | UAP filter, filtrate receiver problems resulted in 8 contamination notices.  |
| March 1967<br>(Riestenberg 1967)               | Plant 8                  | Various   | Sump filter problems; frozen discharge line.   |
| 28 Jun 1967<br>(Levy 1967)                     | Plant 2/3                | 41 to 100 lb. U in 450,000 gal. (17.6 g L <sup>-1</sup> ).                            | Slop Tank F1-25A, located in diked area N of plant, overflowed; most contained, some leaked via trenches to storm sewer.                                     |
| 10 Oct 1968<br>(Starkey 1969)                  | Plant 8                  | 1-2 mR hr <sup>-1</sup> reading   | Liquid material coming from UAP scrubber stack covered ground area of 26' by 15'; no action taken.   |
| 14 Oct 1968<br>(Starkey 1969)                  | Plant 8                  | 5 mR hr <sup>-1</sup> reading   | Spill covering 4' by 4' area at edge of pad near Bldg. 72 scale area; area cleaned.  |
| Feb 1969<br>(Riestenberg 1969a)                | Plant 8                  | About 500 lb. in two weeks  | Trouble with acid filtrate pumps causing low pH readings at lift station; two rebuilt centrifugal pumps installed.   |
| March 1969<br>(Riestenberg 1969b)              | Plant 2/3                | 100 mg L <sup>-1</sup> at 55 -65 gpm in storm sewer                                   | Flushing pad area on west end of Refinery  |
| Dec 1971<br>(Ross 1972)                        | Storm Sewer              | Several hundred lb./mo.   | Should investigate   |
| 27 Apr, 3 and 8 May 1978<br>(Riestenberg 1978) | Plant 6                  | Up to 11 mg L <sup>-1</sup> U at storm sewer; 50 mg L <sup>-1</sup> at MH near Plt 6. | Briquette processing floor leak, and broken storm sewer line; operations stopped until floor repairs completed; flow to MH 175 was diverted to General Sump. |
| 18 January 1988<br>(WMCO 1989)                 | Plant 2/3                | 40 lb. (19 kg) uranium  | Plant 2/3 roof and ground area NE of plant contaminated with uranyl nitrate vented through stack with water vapor.   |
| Spring 1989<br>(Dugan et al. 1990)             | Gravel area S of Plant 7 | 1356 lb. (615 kg) black material; U conc. of 1.0%                                     | Black material (fly ash) fell from a dump truck in the spring; in July, the material was drummed.  |

Because very little rainfall fell during September 1962 (Table L1-23), the loss of uranium to Paddy's Run Creek was only  $6 \pm 3$  kg ( $\pm$  standard deviation) with an estimated monthly volume loss of  $110,000 \pm 21,000$  gallons ( $\pm$  standard deviation). Although highly dependent on rainfall, the average discharge per month to Paddy's Run Creek during this period was roughly 140 kg in 3 to 5 million gallons of water (Tables L-6 and L-7).

In 1955, daily measurements from September through December indicate quite high uranium concentration measurements at MH 175 on November 2 ( $7.6 \text{ mg L}^{-1}$ ), and November 30 ( $6.2 \text{ mg L}^{-1}$ ), compared to an average  $4 \text{ mg U L}^{-1}$ . These events were related to filtration problems in Plant 6, and to cleaning the denitration vapor lines when condensate from the line was sent to the General Sump without analysis; respectively (Chapman 1955). The material was drummed and returned to refinery for further processing.

In February, March, and April 1964, more uranium was lost to the storm sewer (over 5000 lb.) than in any other three-month period of operations (Fischhoff 1964a, 1964b, 1964c). Although no single cause was given for this high loss of materials, varying factors apparently contributed to it. There were extreme weather conditions over the previous eight months with higher than average rainfall. During this time, additional storage pads were being constructed to prevent further spills onto dirt and graveled areas, and this activity may have loosened dirt as a source of contamination in runoff. Finally, work began on repairing the Plant 8 roof where a chronic ground and storm sewer contamination problem existed. During this repair in February and March, all loose contamination was to be removed from the roof before resurfacing and gutter replacement. This loose contamination may have been a source of storm sewer contamination, although it is not clear from the available documentation how the material was handled. This work was completed by April 1964, when a significant portion of the Plant 8 roof area was connected to down spouts directly to the plant sump system (Starkey 1964c). Interestingly, K.N. Ross, of the Industrial Hygiene and Radiation Department who noted contamination problems in memoranda and reports, was on leave from the site at the Nuclear Metals Division in Albany, NY from January 13, 1964 to May 18, 1964.

What seemed to be more common was the situation where a higher than average uranium concentration was noted at MH 175 alerting personnel that an unplanned release or spill of materials containing uranium had occurred. The origin of these higher releases could not always be traced to a definite source or particular location within the facility. For example, in 1960 higher uranium concentrations were measured on January 5 ( $12 \text{ mg L}^{-1}$ ), February 9 ( $10 \text{ mg L}^{-1}$ ), February 18 ( $13 \text{ mg L}^{-1}$ ), April 11 ( $30 \text{ mg L}^{-1}$ ), and May 15 ( $21 \text{ mg L}^{-1}$ ) than the average range of 2 to  $6 \text{ mg L}^{-1}$  uranium (See Table L1-6, annex). Based on these concentration measurements and the corresponding volumes for that day, the probable size of the release or discharge would be calculated (Flowers, 1960a; Beers, 1960b).

On other occasions, situations occurred which did not seem to produce an effect upon the uranium concentration in the effluent at MH 175, such as those in March and April 1961 when an overflow in a sump pit occurred, and Plant 8 UAP acid filtrate spilled and contaminated 40-50 yards of gravel (Table L-10). Furthermore, the addition of contaminated water from extinguishing radioactive fires, or flushing of spill areas into manholes, which were not infrequent events, were not always seen at MH 175 (Fischhoff 1961). Such conditions may have been due to closing the storm sewer near the spill until it was cleaned up, or to an insufficient volume of the effluent for proper flow in the lines.

caused by low rainfall. Another possibility is the occurrence of extreme freezing temperatures during a particular month which would cause accumulation in the lines (Fischhoff 1961). Generally, these latter incidents were noted when melting snow or excess rainfall increased the effluent flow through the lines causing a higher-than-usual flow and greater quantities of uranium at MH 175, such as during periods in February and April 1961 when the average volume and uranium concentration were about twice as high as normal (Table L1-7, annex).

Clearly, Manhole 175 sampling results often did not correlate well with known abnormal releases in the process area. The reverse was also true. In many cases the magnitude of "routine" releases masked the unplanned discharges of some material. On some occasions, excess uranium was noted on the day of an unusual or unplanned occurrence, while other events occurred which did not seem to produce an effect at the sampling location (Fischhoff 1961b). It does appear, however, that the major unplanned releases were detected (e.g. September 10, 1962) at the discharge points from the site. The fact that the large increases in uranium concentration in effluent discharged to the storm sewer system (Figure L-8) correspond to documented accidental spills bears this out. Thus, it is probable that unplanned or accidental liquid releases or spills were detected and have been accounted for as additions to the "usual" or "routine" discharges of uranium measured at Manhole 175 and Paddy's Run Creek. The review of incident reports covering all years of operations suggests that major incidents were not missed, and information regarding major and minor incidents of all kinds were communicated rather frequently by memo, report or letter.

### CHEMICAL AND PHYSICAL FORM OF URANIUM IN LIQUID EFFLUENTS

Several uranium species of both the +4 and +6 oxidation states may have been present in solution in liquid waste streams flowing from the FMPC. The species containing uranium of the +6 oxidation state would probably predominate because most of the uranium discards to the General Sump came from Plant 2/3 (Table L-1), in which the liquid digested material was composed of hexavalent uranium compounds almost exclusively. Uranium in the +4 oxidation state in the form of green salt ( $UF_4$ ) was also discharged from some of the other plants. In addition, some uranium-containing solids which have not been identified specifically were carried in suspension in the liquid waste streams (Alpaugh 1956). There may also have been very small particulates of the insoluble compounds  $U_3O_8$  and  $UO_2$  among the suspended solids.

The species of uranium in the +6 state would include the well-known uranyl ion,  $UO_2^{++}$ , and hydrolytic products such as  $UO_2(OH)^+$ ,  $(UO_2)_2(OH)_2^{++}$ ,  $(UO_2)_3(OH)_4^{++}$ , and others. The very complex hydrolytic reactions involving these species have been described in the literature (Gmelin 1984). The ratios of these various ionic species in waste streams, Paddy's Run Creek, or the Great Miami River would be a function of the pH of the water. Based on the volume of liquid effluent discharged to the river (Tables L-4 and L-7, Figure L-3), most of the  $UF_4$  releases from the plants would have dissolved in the waste streams even though it is not very soluble in water (about 30 mg  $L^{-1}$ ). Hydrolytic reactions of  $UF_4$  probably occurred. Some of the unidentified suspended solids containing uranium that were released in the waste streams might have dissolved during the continued dilution downstream.

The presence of suspended solids in liquid process waste discharged to the Miami River

is considered in assessing the relative solubility of uranium in liquid releases. General concern about the level of total suspended solids (TSS) or filterable materials in the liquid effluents to the river was a long-standing issue at the site (Starkey et al. 1962). The primary problem was that "settleability and radioactivity of the solids are such that the State of Ohio pollution standards cannot always be met without serious curtailment of production processes" (Boback and Heatherton 1958). Daily measurements of TSS were made on 24-hour composite effluent samples at MH 175 beginning in 1956 (NLCO 1956). Table L1-24 in the annex lists the daily measurements of TSS to the river in 1957, and shows the extreme fluctuation for that year because no settling occurred before discharge to the river. The annual average value in 1957 was  $400 \text{ mg L}^{-1}$ , with a maximum of  $4600 \text{ mg L}^{-1}$  measured on October 12, 1957. After April 1958, all solids from the General Sump were sent to Pit 3 for settling, and the liquor pumped to the river via MH 175. This improvement was reflected in the decline of average TSS at MH 175 to less than  $100 \text{ mg L}^{-1}$  in the 1960s and early 1970s (Figure L-10). The decline continued to less than  $25 \text{ mg L}^{-1}$  since 1975. Table L1-25 summarizes the monthly average TSS concentrations in liquid effluents for 1957 to 1966.

Various chemicals and coagulants were tested to assess their effectiveness in removing these solids. In a series of twelve tests in 1958 on effluent samples from around the site, Separan 210, a Dow Chemical Company flocculating agent, reduced the TSS by approximately 70%, beta activity by 90% and alpha activity by 74%. Based on these tests, a TSS concentration of 25 parts per million (ppm) was suggested as a design criterion for wastes released to the river (Boback & Heatherton 1958). After 1958, the TSS in effluents dropped significantly with the transfer of material to the General Sump for settling before release to the river. In the seventies, the U.S. EPA National Pollutant Discharge Elimination System (NPDES) permit for TSS was set at  $100 \text{ mg L}^{-1}$  (Boback et al. 1977).

Similarly, methods were assessed for their usefulness in removing soluble uranium from the liquid effluent (Dugan 1971). In 1971, tests results showed that the addition of lime slurry decreased the soluble uranium concentration of storm sewer effluent. However, the addition of lime to the storm sewer to neutralize acid spills and to prevent corrosion at the lift station was usually associated with higher TSS levels in effluents to the river (Boback 1971b). Other causes of TSS exceeding the limit were related to runoff from the coal pile (Starkey 1968b) and variable pH of the effluent (Boback 1971c).

In summary, the ratios of various ionic species of uranium compounds in waste streams, Paddy's Run Creek, or the Great Miami River is a function of the pH of the water. Based on the high volume of liquid effluent released, many of the uranium species released from the plants would have dissolved in the waste streams, although suspended solids were prevalent in the effluent. Among the suspended solids may have been very small particulates of the insoluble compounds  $\text{U}_3\text{O}_8$  and  $\text{UO}_2$ . It is clear that not all the suspended solids measured on a daily basis were uranium, but the average monthly values may provide an upper bound for the amount of insoluble uranium released in liquid effluent.

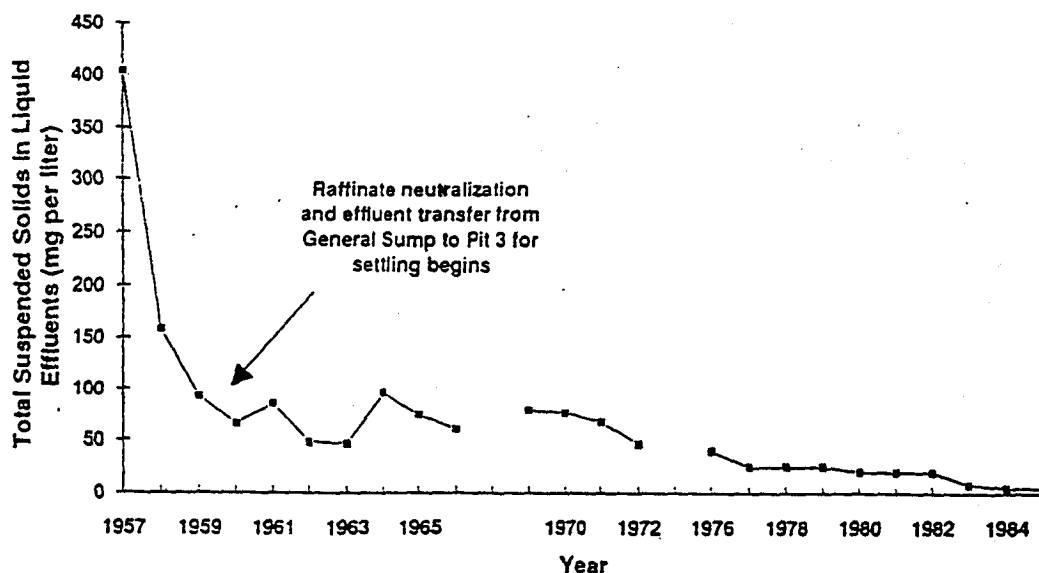


Figure L-10. Annual average concentration of total suspended solids in liquid effluents released at MH 175 to the river. Daily measurements were made beginning in 1956 at MH 175. Major improvements in the liquid effluent treatment in 1958 lead to a significant decrease in TSS.

## RADIONUCLIDES OTHER THAN URANIUM

Uranium was the primary material processed at the FMPC with some thorium processing occurring at various times. Most of the feed material had previously been separated chemically from the naturally occurring daughter radionuclides. Consequently, most effluents from the facility consisted primarily of uranium and, when it was being processed, thorium. Beginning in 1953, thorium operations were performed in the Metals Fabrication Plant (Plant 6), Recovery Plant (Plant 8), Special Products Plant (Plant 9) and the Pilot Plant. Thorium oxide for thorium metal conversion was made during the period of 1954 to 1956 by aqueous precipitation of thorium fluoride from an aqueous hydrofluoric acid solution (Jester 1964). Severe corrosion problems, caused by hot nitric-hydrofluoric acid mixtures, forced a change to the more expensive oxalate process in Plant 9. Appendix D in this report, and Appendix C in the Task 4 report, Environmental Pathways — Models and Validation, describe the products of radioactive decay of uranium and thorium. Four isotopes of radium naturally occur as decay products of thorium and uranium. Two of these,  $^{228}\text{Ra}$  and  $^{224}\text{Ra}$ , are decay products of thorium. Radium-223 is a decay product of  $^{235}\text{U}$ , and  $^{226}\text{Ra}$  is a decay product of  $^{238}\text{U}$ . When the relative importance of releases of these radionuclides to water was assessed for the 1960 to 1962 period, it was found that the radium isotopes were of primary importance (Appendix C, Killough et al. 1993).

Appendix D also describes other radionuclides that were released during FMPC operations from the processing of recycled uranium, that is, uranium that was not completely separated from fission and activation products before it was returned to the FMPC as feed material. Recycled uranium was processed at the FMPC beginning in the fall of 1962 (Voillequé et al. 1991). When recycled uranium was processed, some fission and

activation products were discharged from the site in both liquid and airborne effluents. This section provides annual estimates of these radionuclides released in liquid waste from the site. Table L-11 lists these products, information on release and measurement periods, and sources of information used to generate the source terms.

Table L-11. Decay, Fission and Activation Products Released in Liquid Effluents From the FMPC

| Materials in Liquid Effluents | Releases Began | Measurements Began | Information Source |
|-------------------------------|----------------|--------------------|--------------------|
| <b>Decay Products</b>         |                |                    |                    |
| Total Thorium                 | 1954           | 1956               | a, b, c            |
| Total Radium                  | 1952           | 1955               | a                  |
| $^{226}\text{Ra}$             | 1952           | 1968               | a, b, c            |
| $^{228}\text{Ra}$             | 1954           | 1968               | a, b, c            |
| <b>Fission Products</b>       |                |                    |                    |
| $^{90}\text{Sr}$              | Fall 1962      | 1976               | d                  |
| $^{99}\text{Tc}$              | Fall 1962      | 1969               | b, d               |
| $^{106}\text{Ru}$             | Fall 1962      | 1976               | b, d               |
| $^{137}\text{Cs}$             | Fall 1962      | 1976               | d                  |
| <b>Activation Products</b>    |                |                    |                    |
| $^{237}\text{Np}$             | Fall 1962      | 1976               | d                  |
| $^{238}\text{Pu}$             | Fall 1962      | 1976               | d                  |
| $^{239,240}\text{Pu}$         | Fall 1962      | 1976               | d                  |

<sup>a</sup> Original analytical data sheets for some periods; NLCO 1955b, NLCO 1956, NLCO 1957, NLCO 1969, NLCO 1970, NLCO 1971, NLCO 1972, NLCO 1973, NLCO 1974.

<sup>b</sup> Various monthly reports including routine operating loss reports, Industrial Hygiene and Radiation Department reports and Aquifer Contamination Reports.

<sup>c</sup> Based on correlations between releases of uranium and other radionuclides when measurements were made; see Table L-13.

<sup>d</sup> Based on correlations between releases of uranium and other radionuclides when measurements were made; see Table L-12.

Thorium and radium were measured in liquid effluents beginning in the early 1950s, and original analytical data sheets for radium measurements were located for 1955, 1956, 1957, 1969 and 1970-1974 (Tables L1-25 to L1-32), and for thorium for 1956 and 1957 (Tables L1-33 and L1-34). Measurements were made on weekly or biweekly composites for radium, and monthly composites for thorium. A regular sampling program for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  was begun in 1968, for  $^{99}\text{Tc}$  in 1969, and for all other radionuclides of interest in 1976 (Boback et al. 1987, NLCO 1975). Periodic monthly composite samples from MH 175 were analyzed for  $^{99}\text{Tc}$  (technetium) and  $^{106}\text{Ru}$ - $^{106}\text{Rh}$  (ruthenium-rhodium) activity beginning in the late 1960s when higher levels of beta activity were measured in effluents sent to Waste Pit 3 (Starkey 1968a, NLCO 1971, NLCO 1974). However, the bioassay lab procedure for  $^{106}\text{Ru}$  was not documented for those years (Berger et al. 1985). Routinely, monthly composites of the daily samples from MH 175 were analyzed for  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ,  $^{106}\text{Ru}$  and thorium with annual composites analyzed for the other radionuclides through the mid-1980s. Analysis of  $^{232}\text{Th}$  in liquid wastes to the river replaced total thorium measurements in 1984 (Facemire et al. 1985).

Release estimates of these other radionuclides are based on correlations between the total annual releases of uranium and those of the other radionuclides. These ratios of releases, computed for years when measurements were made, provide a basis for estimating the release of the other radionuclides for years when they were not measured. This methodology is described in Appendix D in the present report, and in Appendix C of Task 4 (Killough et al. 1993). Ratios of the annual average activity of a radionuclide (or, quantity of thorium) to the annual uranium quantity were calculated for years when data were available. The measured concentrations at MH 175 reported in analytical data sheets were used to calculate the ratio for some years (NLCO 1955b, NLCO 1956, NLCO 1957, NLCO 1969, NLCO 1970, NLCO 1971, NLCO 1972, NLCO 1973, NLCO 1974). Annual average concentrations of radium, thorium and the fission and activation products in liquid effluents were reported by the FMPC in historic release reports (Boback et al. 1987), and in annual environmental monitoring reports (Boback et al. 1977, Boback et al. 1978, Boback & Ross 1979, Boback & Ross 1980, Boback & Ross 1981, Fleming et al. 1982, Fleming & Ross 1983, Fleming & Ross 1984, Facemire et al. 1985, Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989). The annual average uranium concentration at MH 175, or total quantity of uranium to the river was used for these correlations depending upon the source of data (analytical data sheets or total release estimates, respectively). The variability of the release ratio from year to year was considered in deriving the uncertainty associated with the estimated releases of these other radionuclides. The release estimates and uncertainty analysis were computed using Monte Carlo techniques in the Crystal Ball® program (Decisioneering 1993), assuming a lognormal distribution for the ratios of the radionuclide of interest to uranium.

Table L-12 shows the relative concentrations of activation and fission products relative to uranium,  $\mu\text{Ci (kg U)}^{-1}$ , based on thirteen years of measurements. For radium and thorium, the ratios are based on a somewhat longer measurement history. Table L-13 shows that the ratios of releases are based on measurements as early as 1956 for thorium, and 1968 for  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$ . Measurements of total radium, made in the early 1950s (NLCO 1955b, NLCO 1956, NLCO 1957), were used to calculate a ratio of  $^{226}\text{Ra}$  activity (assuming a specific activity of  $0.99 \mu\text{Ci per } \mu\text{g Ra}$ ) to uranium, which was used to estimate  $^{226}\text{Ra}$  releases in the 1950s. During the 1950s, the  $^{226}\text{Ra}$  concentrations are higher than in later years because, from October 1955 to August 1958, some of the uranium ore processed was pitchblende, which had very high uranium (and thus decay product) concentrations (See Appendix J). For later years, a second  $^{226}\text{Ra}$  ratio ( $50 \pm 80 \mu\text{Ci (kg U)}^{-1}$ ), based on measurements made from 1968-1988, was used to calculate releases estimates. A single ratio for  $^{228}\text{Ra}$  to uranium ( $90 \pm 80 \mu\text{Ci (kg U)}^{-1}$ ), based on measurements made from 1968-1988, was used to calculate  $^{228}\text{Ra}$  releases. These estimates were calculated for years when thorium processing occurred, because  $^{228}\text{Ra}$  is a decay product of thorium (See Appendix D).

Relative concentrations of thorium with respect to uranium are reported as kilograms of thorium per kilogram of uranium,  $(\text{kg Th}) (\text{kg U})^{-1}$ . Because thorium processing occurred only during specific years, release estimates are calculated for 1954 to 1957, and for 1968-1988. Ratios of thorium to uranium quantities were calculated for two periods: the 1950s and 1964-1988. The ratio for the early time [ $0.41 \pm 0.04 \text{ kg Th (kg U)}^{-1}$ ] is based on concentrations of thorium to uranium measured at MH 175 in 1956 and 1957 (NLCO 1956, NLCO 1957). For later years, the ratio [ $0.013 \pm 0.015 \text{ kg Th (kg U)}^{-1}$ ] is based on measurements from 1967-1988.



Table L-12. Relative Concentrations of Activation (Pu, Np) and Fission Products ( $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$ ) Measured in Liquid Waste Discharges,  $\mu\text{Ci (kg U)}^{-1}$  <sup>a</sup>

| Year   | $^{239,240}\text{Pu}$ | $^{238}\text{Pu}$ | $^{237}\text{Np}$ | $^{137}\text{Cs}$ | $^{106}\text{Ru}$ | $^{99}\text{Tc}$  | $^{90}\text{Sr}$ |
|--------|-----------------------|-------------------|-------------------|-------------------|-------------------|-------------------|------------------|
| 1976   | 0.00024               | 0.00049           | 0.00024           | 24                | 3.7               | $1.1 \times 10^4$ | no data          |
| 1977   | <0.053                | <0.024            | <0.48             | 80                | 7.8               | $9.5 \times 10^1$ | 71               |
| 1978   | <0.038                | <0.027            | 0.036             | 17                | 1.2               | $1.1 \times 10^2$ | 7.8              |
| 1979   | 0.024                 | 0.0082            | 0.16              | 5                 | 1.5               | $2.8 \times 10^3$ | 2.6              |
| 1980   | 2.2                   | 0.006             | <0.16             | 16                | 1.4               | $1.4 \times 10^3$ | 4.1              |
| 1981   | 0.05                  | 0.0088            | <0.24             | 4                 | 1.2               | $7.3 \times 10^3$ | 4.3              |
| 1982   | 0.02                  | 0.0065            | 0.4               | 3.7               | 0.045             | $1.3 \times 10^4$ | 4.2              |
| 1983   | 0.13                  | 0.0085            | <0.30             | 9.3               | 0.51              | $3.5 \times 10^4$ | 9.8              |
| 1984   | 0.049                 | 0.029             | 0.20              | 17                | 0.49              | $1.9 \times 10^4$ | 12               |
| 1985   | 0.024                 | 0.012             | <0.27             | 16                | <0.68             | $1.3 \times 10^4$ | 8.5              |
| 1986   | <0.022                | <0.022            | <0.022            | <2.2              | <22               | $3.3 \times 10^3$ | 2                |
| 1987   | <0.073                | <0.072            | <0.31             | <9.7              | <43               | $3.5 \times 10^3$ | 2.9              |
| 1988   | <0.028                | <0.02             | <0.04             | <6                | <39               | $7.3 \times 10^3$ | 1.5              |
| Mean   | 0.31                  | 0.01              | 0.16              | 19                | 2.0               | $8.9 \times 10^3$ | 11               |
| StdDev | 0.76                  | 0.01              | 0.16              | 22                | 2.4               | $9.7 \times 10^3$ | 19               |

<sup>a</sup> Data for these ratios of activity ( $\mu\text{Ci}$ ) to quantity ( $\text{kg}$ ) of uranium are taken from Annual Environmental Monitoring Reports (Boback et al. 1977, Boback et al. 1978, Boback & Ross 1979, Boback & Ross 1980, Boback & Ross 1981, Fleming et al. 1982, Fleming & Ross 1983, Fleming & Ross 1984, Facemire et al. 1985, Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989).

The result of these computations for thorium are shown in Figure L-11, where the relative quantities of total thorium are compared to the total quantity of uranium discharged in liquid effluents for those years when thorium was processed. The higher thorium releases in the 1950s were related to the fact that thorium oxide for thorium metal conversion was made during the period of 1954 to 1956 by aqueous precipitation of thorium fluoride from an aqueous hydrofluoric acid solution (Jester 1964). This process caused severe corrosion problems, caused by hot nitric-hydrofluoric acid mixtures. For later thorium operations, a change to the more expensive oxalate process in Plant 9 occurred. After 1964, the quantities of thorium discharged to the river were approximately two orders of magnitude less than the quantities of uranium. The thorium releases in the mid-1950s were substantially higher. Similarly, the relative changes in activity of  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$  in liquid effluents from the FMPC with time, shown in Figure L-12, are similar to the pattern of thorium releases. The highest releases occurred during the 1950s and 1960s, with a gradual decrease in activity in the 1970s and 1980s. Tables L-14 and L-15 show the annual estimates for thorium,  $^{228}\text{Ra}$ , and  $^{226}\text{Ra}$ , discharged in liquid effluents from the FMPC, along with the uncertainty estimates for each measurement.

Figure L-13 displays the total release estimates for the radionuclides,  $^{239,240}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{237}\text{Np}$ ,  $^{137}\text{Cs}$ ,  $^{106}\text{Ru}$ ,  $^{99}\text{Tc}$ , and  $^{90}\text{Sr}$ , for all years of operations. Table L-16 provides the annual estimates of fission and activation products discharged in liquid effluents from the FMPC for each year from 1962 to 1988. Because the processing of recycled uranium at the

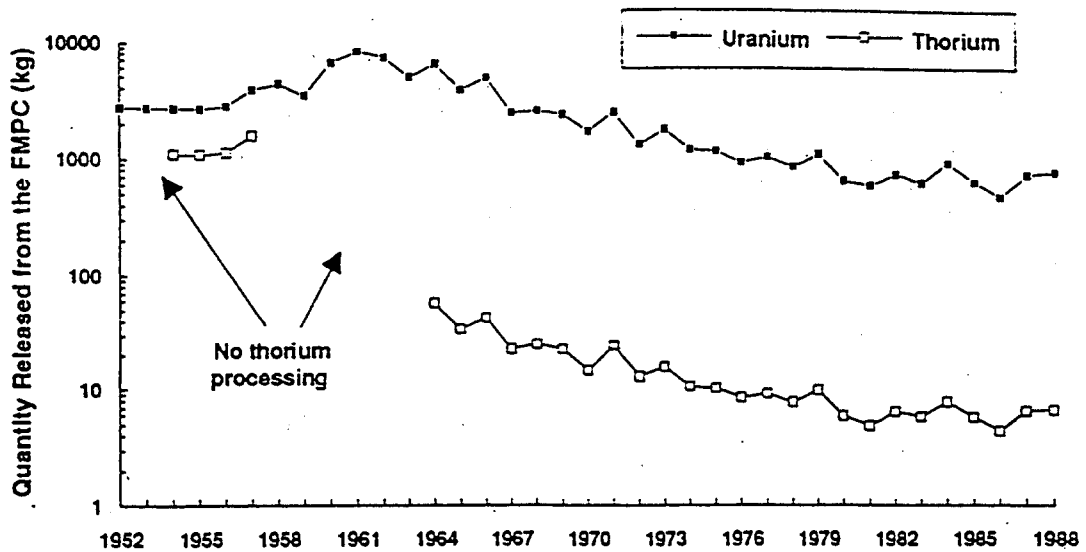
FMPC did not begin until October 1962, values for 1962 are based on only three months of operation. Since 1962, total releases of  $^{99}\text{Tc}$  were approximately 300,000 mCi (300 Ci), with an uncertainty range of 100,000 to 800,000 mCi (100 to 800 Ci). The best estimate for releases of  $^{239,240}\text{Pu}$  since 1962 is 8.8 mCi, with an uncertainty range of 1.9 to about 30 mCi.

Table L-13. Relative Concentrations of Radium and Thorium to Uranium Measured in Liquid Waste Discharges<sup>a</sup>

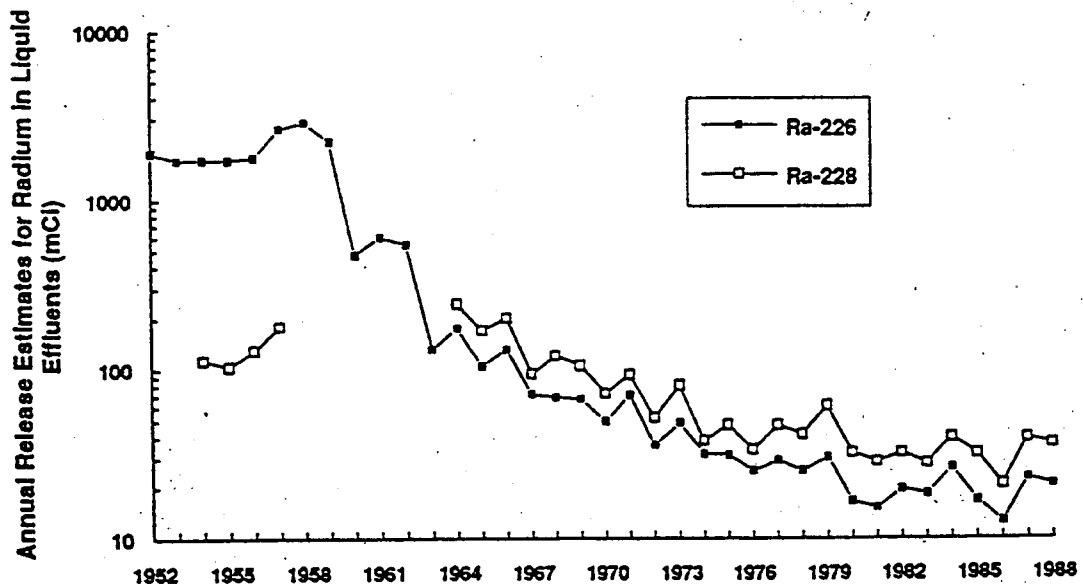
| Year          | $^{226}\text{Ra}$                 | $^{228}\text{Ra}$                            | Thorium                           |
|---------------|-----------------------------------|--|-----------------------------------|
|               | $\mu\text{Ci}/(\text{kg U})^{-1}$ | $\mu\text{Ci}/(\text{kg U})^{-1} \text{ }^a$ | $\text{kg Th}/(\text{kg U})^{-1}$ |
| 1955          | 1600 <sup>b</sup>                 |  |                                   |
| 1956          | 220 <sup>b</sup>                  |  | 0.44                              |
| 1957          | 530 <sup>b</sup>                  |  | 0.37                              |
| Mean (1950s)  | 780                               |  | 0.41                              |
| Stdev (1950s) | 590                               |  | 0.04                              |
| 1967          |                                   |  | 0.012                             |
| 1968          | 270                               | 590  | 0.069                             |
| 1969          | 250                               | 390  | 0.028                             |
| 1970          | 104                               | 260  | 0.015                             |
| 1971          | 61                                | 24   | 0.018                             |
| 1972          | 48                                | 13   | 0.016                             |
| 1973          | 21                                | 5.30   | 0.008                             |
| 1974          | 7.50                              | 5.60   | 0.017                             |
| 1975          | 7.02                              | 8.60   | 0.0035                            |
| 1976          | 9.72                              | 11   | 0.0076                            |
| 1977          | 8.00                              | 77   | 0.0057                            |
| 1978          | 3.81                              | 5.10   | 0.0065                            |
| 1979          | 0.68                              | 8.20   | 0.0061                            |
| 1980          | 0.56                              | 5.20   | 0.0033                            |
| 1981          | 19                                | 12   | 0.0053                            |
| 1982          | 4.03                              | 17   | 0.0052                            |
| 1983          | 2.40                              | 11   | 0.0035                            |
| 1984          | <17                               | <14  | 0.0044                            |
| Mean          | 50                                | 90   | 0.013                             |
| Stdev         | 80                                | 170  | 0.015                             |

<sup>a</sup> Values are derived from the following sources: routine analytical data sheets for uranium,  $^{226}\text{Ra}$  and thorium in the 1950s (see Tables L1-1 to L1-13, L1-26 to L1-28 and L1-34 and L1-36), and  $^{228}\text{Ra}$  in 1969, 1967-1975, Boback et al. 1987; 1976-1988, Annual Environmental Monitoring Reports (Boback et al. 1977, Boback et al. 1978, Boback & Ross 1979, Boback & Ross 1980, Boback & Ross 1981, Fleming et al. 1982, Fleming & Ross 1983, Fleming & Ross 1984, Facemire et al. 1985, Aas et al. 1986, Aas et al. 1987, WMCO 1988, WMCO 1989).

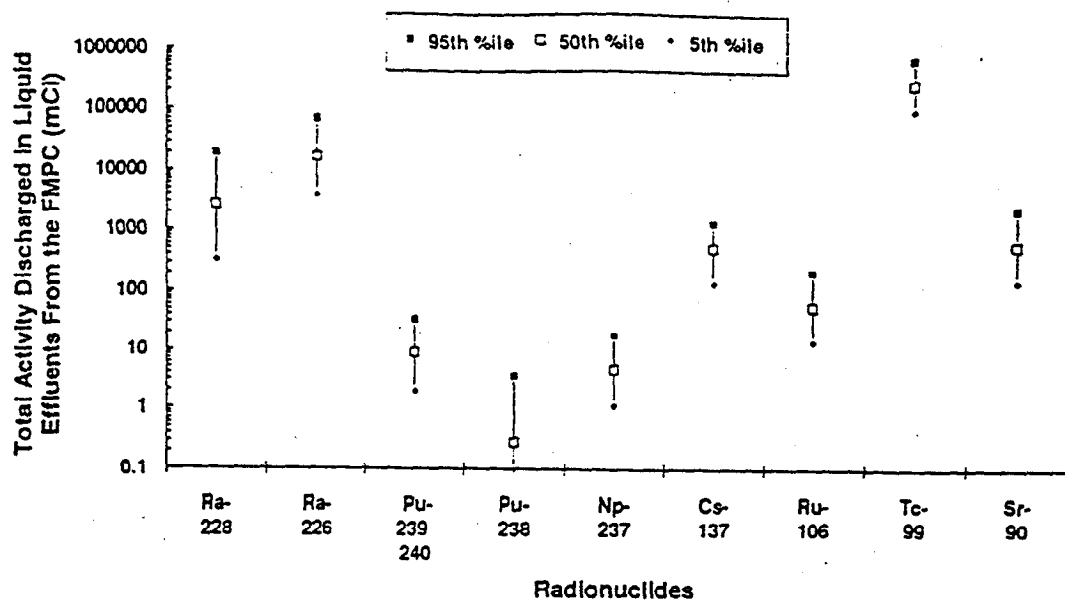
<sup>b</sup> For 1955, 1956 and 1957, the ratio is derived from total radium measurements of  $\mu\text{g}$  per mL (Tables L1-26 to L1-28), assuming a specific activity of 0.99  $\mu\text{Ci}$  per  $\mu\text{g}$  Ra.



**Figure L-11.** Relative annual estimates for uranium and thorium released in liquid effluents from the FMPC. Thorium processing occurred from 1954 to 1957 and from 1964 through 1988. The uranium values represent total uranium quantities released to both the Great Miami River and to Paddy's Run Creek. Figure L-2 shows the uranium releases individually to the river and to Paddy's Run.



**Figure L-12.** Annual estimates of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  releases in liquid effluents from the FMPC. Release estimates for  $^{228}\text{Ra}$ , a decay product of thorium, are given for 1954-1957, and 1964-1988, the years when thorium processing occurred (see Table L-15).



**Figure L-13.** Estimates of total activity of radium, fission and activation products released from the FMPC in liquid effluents. For radium, the values represent releases from 1952 to 1988; for the other radionuclides, releases occurred from 1962 onward. The uncertainty of each estimate is shown as the 95th and 5th percentiles.

Tables L-14 to L-16 also show the gradual decrease in release estimates in the 1970s and 1980s. These decreases over time for all radioactive materials are related to a general reduction in production activities from the higher levels observed in the fifties and sixties, as well as to a number of changes in liquid effluent handling and treatment at the site, including

- major improvements in the General Sump area for waste effluent processing in 1968, and the
- construction of new wet chemical Waste Pit 5 by 1969.

By 1967, Waste Pit 3 was nearly at its capacity. At the same time, the General Sump was processing large volumes of soluble high beta activity material from a variety of processing campaigns. However, the General Sump was in more frequent need of repairs by the mid-1960s. When holding tanks in the General Sump were being repaired, virtually all effluent from the General Sump was pumped to Waste Pit 3 before proper precipitation and settling could occur. To make more room in the pit, pumping from the waste pit clearwell was increased prior to complete settling of the material. A consequence of this was higher discharges of radionuclides to the river during the sixties.

In 1969, two new 15,000 gallon and a new 50,000 gallon sludge settling tanks in the General Sump area were installed, and a new head tank for regulating continuous discharge to the river was operational (OHIO 1968). Most importantly, the construction of the new wet chemical pit began on July 15, 1968, and was receiving material by the end of that year (Starkey 1968c). The first effluent from the new pit was pumped to the river on January 6, 1969 (Starkey 1969a).

**Table L-14. Annual Estimates of Thorium Discharged in Liquid Effluents From the FMPC (kg) <sup>a</sup>**

| Year            | Median Estimate | 5th %ile | 95th %ile |
|-----------------|-----------------|----------|-----------|
| 1954            | 1100            | 800      | 1500      |
| 1955            | 1100            | 830      | 1400      |
| 1956            | 1200            | 910      | 1500      |
| 1957            | 1600            | 1300     | 2100      |
| 1964            | 58              | 11       | 280       |
| 1965            | 34              | 8        | 150       |
| 1966            | 43              | 9        | 190       |
| 1967            | 22              | 5        | 100       |
| 1968            | 24              | 5        | 110       |
| 1969            | 22              | 5        | 110       |
| 1970            | 14              | 3        | 63        |
| 1971            | 24              | 5        | 110       |
| 1972            | 13              | 3        | 50        |
| 1973            | 16              | 3        | 67        |
| 1974            | 11              | 2        | 48        |
| 1975            | 10              | 2        | 48        |
| 1976            | 9               | 2        | 43        |
| 1977            | 9               | 2        | 44        |
| 1978            | 8               | 2        | 41        |
| 1979            | 10              | 2        | 51        |
| 1980            | 6               | 1        | 28        |
| 1981            | 5               | 1        | 24        |
| 1982            | 7               | 1        | 30        |
| 1983            | 6               | 1        | 26        |
| 1984            | 8               | 2        | 37        |
| 1985            | 6               | 1        | 25        |
| 1986            | 4               | 1        | 20        |
| 1987            | 7               | 1        | 33        |
| 1988            | 7               | 1        | 35        |
| Total all years | 5800            | 3800     | 9400      |

<sup>a</sup> Estimates and uncertainties were calculated with CrystalBall Version 3.0 (Decisioneering 1993). No thorium processing occurred in 1952, 1953, or 1958-1963 (see Appendix C).

In the sixties, unusually high soluble beta activity, measured in the General Sump and the waste pits, was attributed to <sup>106</sup>Ru and <sup>99</sup>Tc from various processing campaigns such as the processing of NFS feed material which contained <sup>106</sup>Ru, or to high <sup>236</sup>U refinery runs (Starkey 1967b). In the oxidized state, both are soluble in basic and acidic solutions, so that they were not effectively removed by passage through the General Sump. By 1970, the beta activity attributed to soluble <sup>106</sup>Ru and <sup>99</sup>Tc had gradually decreased from the levels seen previously (Boback 1969).

Table L-15. Annual Estimates of  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$  Discharged in  
Liquid Effluents From the FMPC (mCi)<sup>a</sup>

| Year   | Radium-228         |          |           | Radium-226         |          |           |
|--------|--------------------|----------|-----------|--------------------|----------|-----------|
|        | Median<br>Estimate | 5th %ile | 95th %ile | Median<br>Estimate | 5th %ile | 95th %ile |
| 1952   |                    |          |           | 1900               | 616      | 5300      |
| 1953   |                    |          |           | 1700               | 535      | 4800      |
| 1954   | 110                | 14       | 930       | 1700               | 584      | 5300      |
| 1955   | 100                | 12       | 710       | 1700               | 622      | 5200      |
| 1956   | 130                | 17       | 1200      | 1800               | 623      | 5400      |
| 1957   | 180                | 25       | 1300      | 2600               | 907      | 7700      |
| 1958   |                    |          |           | 2900               | 1105     | 8500      |
| 1959   |                    |          |           | 2200               | 822      | 6400      |
| 1960   |                    |          |           | 480                | 46       | 3300      |
| 1961   |                    |          |           | 600                | 54       | 6300      |
| 1962   |                    |          |           | 540                | 52       | 5400      |
| 1963   |                    |          |           | 130                | 21       | 870       |
| 1964   | 250                | 36       | 2000      | 180                | 27       | 1100      |
| 1965   | 170                | 23       | 1400      | 110                | 17       | 680       |
| 1966   | 200                | 25       | 1600      | 130                | 21       | 830       |
| 1967   | 96                 | 13       | 820       | 72                 | 10       | 460       |
| 1968   | 120                | 15       | 1050      | 69                 | 13       | 460       |
| 1969   | 110                | 11       | 880       | 68                 | 11       | 490       |
| 1970   | 74                 | 9        | 670       | 50                 | 7        | 320       |
| 1971   | 95                 | 13       | 800       | 72                 | 12       | 500       |
| 1972   | 53                 | 7        | 450       | 36                 | 5        | 240       |
| 1973   | 82                 | 10       | 690       | 49                 | 8        | 360       |
| 1974   | 38                 | 5        | 340       | 32                 | 5        | 200       |
| 1975   | 48                 | 6        | 440       | 31                 | 5        | 200       |
| 1976   | 34                 | 4        | 320       | 25                 | 4        | 170       |
| 1977   | 48                 | 6        | 370       | 29                 | 5        | 179       |
| 1978   | 42                 | 5        | 300       | 25                 | 4        | 150       |
| 1979   | 63                 | 6        | 500       | 31                 | 4        | 200       |
| 1980   | 33                 | 4        | 250       | 17                 | 3        | 120       |
| 1981   | 29                 | 4        | 280       | 15                 | 2        | 100       |
| 1982   | 33                 | 5        | 260       | 20                 | 3        | 120       |
| 1983   | 28                 | 4        | 250       | 18                 | 2        | 110       |
| 1984   | 41                 | 5        | 370       | 27                 | 4        | 180       |
| 1985   | 32                 | 4        | 280       | 17                 | 3        | 100       |
| 1986   | 21                 | 3        | 180       | 13                 | 2        | 83        |
| 1987   | 40                 | 5        | 300       | 23                 | 3        | 130       |
| 1988   | 37                 | 4        | 260       | 21                 | 3        | 140       |
| Totals | 2700               | 330      | 20000     | 18000              | 15000    | 22000     |

<sup>a</sup> Estimates and uncertainties were calculated with CrystalBall Version 3.0 (Decisioneering 1993).

<sup>b</sup> Radium-228 is a decay product of thorium; estimates of  $^{228}\text{Ra}$  releases are given for 1954-1957, and 1964-1988, the years when thorium processing occurred.

Table L-16. Annual Activity Estimates of Fission and Activation Products  
Discharged in Liquid Effluents From the FMPC (mCi)<sup>a</sup>

| Year <sup>b</sup> | <sup>239,240</sup> Pu | <sup>238</sup> Pu | <sup>237</sup> Np | <sup>137</sup> Cs | <sup>106</sup> Ru | <sup>99</sup> Tc | <sup>90</sup> Sr |
|-------------------|-----------------------|-------------------|-------------------|-------------------|-------------------|------------------|------------------|
| 1962              | 0.39                  | 0.01              | 0.21              | 25                | 2.6               | 11000            | 270              |
| 1963              | 1.29                  | 0.04              | 0.69              | 82                | 8.6               | 38000            | 900              |
| 1964              | 1.23                  | 0.04              | 0.66              | 78                | 8.2               | 36000            | 860              |
| 1965              | 0.87                  | 0.03              | 0.46              | 55                | 5.8               | 26000            | 610              |
| 1966              | 1.35                  | 0.05              | 0.72              | 86                | 9.0               | 39600            | 940              |
| 1967              | 0.57                  | 0.02              | 0.30              | 36                | 3.8               | 16600            | 390              |
| 1968              | 0.72                  | 0.02              | 0.38              | 46                | 4.8               | 21000            | 500              |
| 1969              | 0.69                  | 0.02              | 0.37              | 44                | 4.6               | 20200            | 480              |
| 1970              | 0.45                  | 0.02              | 0.24              | 29                | 3.0               | 13000            | 310              |
| 1971              | 0.66                  | 0.02              | 0.35              | 42                | 4.4               | 19000            | 460              |
| 1972              | 0.33                  | 0.01              | 0.18              | 21                | 2.2               | 9700             | 230              |
| 1973              | 0.51                  | 0.02              | 0.27              | 32                | 3.4               | 15000            | 360              |
| 1974              | 0.22                  | 0.01              | 0.12              | 14                | 1.4               | 6300             | 150              |
| 1975              | 0.30                  | 0.01              | 0.16              | 19                | 2.0               | 8900             | 210              |
| 1976              | 0.22                  | 0.01              | 0.12              | 14                | 1.5               | 6400             | 150              |
| 1977              | 0.27                  | 0.01              | 0.15              | 17                | 1.8               | 8000             | 190              |
| 1978              | 0.26                  | 0.01              | 0.14              | 16                | 1.7               | 7500             | 180              |
| 1979              | 0.32                  | 0.01              | 0.17              | 20                | 2.1               | 9200             | 220              |
| 1980              | 0.19                  | 0.01              | 0.10              | 12                | 1.3               | 5600             | 130              |
| 1981              | 0.18                  | 0.01              | 0.10              | 11                | 1.2               | 5300             | 130              |
| 1982              | 0.23                  | 0.01              | 0.12              | 14                | 1.5               | 6600             | 160              |
| 1983              | 0.18                  | 0.01              | 0.09              | 11                | 1.2               | 5200             | 120              |
| 1984              | 0.27                  | 0.01              | 0.14              | 17                | 1.8               | 7900             | 190              |
| 1985              | 0.17                  | 0.01              | 0.09              | 10                | 1.1               | 4800             | 120              |
| 1986              | 0.13                  | 0.00              | 0.07              | 8                 | 0.84              | 3700             | 88               |
| 1987              | 0.21                  | 0.01              | 0.11              | 13                | 1.4               | 6200             | 150              |
| 1988              | 0.22                  | 0.01              | 0.12              | 14                | 1.5               | 6500             | 160              |
| Total: all years  | 8.8                   | 0.28              | 4.4               | 540               | 56                | 300000           | 600              |
| (5th-95th %ile)   | (1.9-33)              | (0.16-3.4)        | (1.1-18)          | (140-1900)        | (14-220)          | (110000-800000)  | (1500-24000)     |

<sup>a</sup> The median estimates are based on the average ratio of measured activity of these radionuclides to the quantity of uranium released in liquid effluent from 1976 onward. The values are reported in millicuries (mCi); one mCi is equal to 1000 microcuries (μCi) or 0.001 curie (Ci).

<sup>b</sup> Processing of recycled uranium at the FMPC did not begin until October 1962. Consequently, values for 1962 are based on only three months of processing.

By 1969, when the average concentration of <sup>228</sup>Ra in the plant effluent was about 1.8 disintegrations per minute per milliliter (d/m/mL) (Table L1-29, annex), the Oak Ridge Operations Atomic Energy Commission requested the concentration of <sup>228</sup>Ra in the wastes discharged to the river be reduced (Boback 1969). At that time, the Pilot Plant thorium extraction process was the major source of this radionuclide. A barium sulfate precipitation at the Pilot Plant and additional treatment at the General Sump were intended to reduce

the  $^{228}\text{Ra}$  in the extraction waste stream before being pumped to Waste Pit 5. Beginning the oxalate process for thorium recovery in Plant 8 in 1969, however, prevented lowering the concentrations quickly. By mid-1970, work at the General Sump had increased as a result of processing thorium scrap in Plant 8. The clear liquid from this process was pumped to the Chemical Waste Pit 5 and the solids were reprocessed through Plant 8. The reduction in average  $^{228}\text{Ra}$  concentration at MH 175 from 3.2 d/m/mL in December 1969 to 1.6 d/m/mL in March 1970 (Table L1-29) occurred when there were no thorium extraction operations in the Pilot Plant during that period. Even though all thorium effluent from both Plant 8 and the Pilot Plant was pumped to Pit 5,  $^{228}\text{Ra}$  in the effluent from the General Sump to the river still averaged 5.0 d/m/mL in August 1970, and was attributed to incoming effluents from various plants (Boback 1970). By the end of 1970, the concentration of  $^{228}\text{Ra}$  had declined. In 1971, the General Sump began solidifying certain  $^{228}\text{Ra}$ -bearing wastes from Plant 8 for shipment and burial offsite (Pennack 1971).

## SUMMARY AND CONCLUSIONS

Liquid wastes at the FMPC came from three main sources: (1) process water from the production area via the General Sump and clearwell portion of the waste pits, (2) from the sanitary sewer treatment plant, and (3) from the storm sewer system. The facilities for handling liquid wastes from the process areas included collection sumps and treatment equipment at each plant to remove uranium from process waste water before it was pumped to the General Sump. From the General Sump, the effluent was pumped to the waste pits where settling occurred, after which the liquid was decanted to the clearwell portion of the pit. Key improvements in the liquid handling system at the FMPC, especially in 1958, 1968 and 1985, were reflected in noticeable declines in concentrations of uranium, thorium and other radionuclides, as well as in total suspended solids measured at the discharge point to the river.

Liquid effluent left the FMPC site at two locations. The main pipeline exited via Manhole 175 (MH 175) into the Great Miami River at a point almost directly east of the plant site. Liquid waste water also left the site via the storm sewer outfall ditch and runoff into Paddy's Run Creek, when the storm sewer lift station could not handle the runoff volume. Effluent volume and total uranium concentration were measured routinely at both locations (MH 175 and the storm sewer outfall ditch). Daily analytical data sheets, and monthly reports of effluent volume and uranium discharged form the basis of our source term computations.

Table L-17 summarizes our estimates for releases of materials in liquid effluents from the FMPC for all years of operation. Our best estimate of uranium released to the Great Miami River for all years is 82,000 kg. The 5th to 95th percentile uncertainty range is 71,000 to 94,000 kg of uranium. The sources of uncertainty for losses through MH 175 to the Great Miami River come primarily from the analytical errors in the daily measurements of water flow, and in sampling and determination of uranium concentration in the water. Some estimates of uranium in liquid wastes have been made by others on an annual basis (Boback 1971a), or in summary reports evaluating the past discharge history of the facility (Rathgens 1977, Boback et al., 1985). These estimates of uranium to surface water from 1951 through 1984 range from 74,000 to 77,000 kg (Boback et al. 1987, Galper 1988) and fall



within the uncertainty range of our estimates. Revisions to historic discharge reports generally focused on amending estimates of uranium loss to airborne effluents, and did not include updated figures for liquid effluents (Boback et al. 1985, Boback et al. 1987).

**Table L-17. Summary of Total Estimates of Radioactive Materials Released From the FMPC in Liquid Effluents For All Years of Operation**

| Material Released to Great Miami River | Median Value  | Uncertainty Range (5th %ile to 95th %ile) |
|--|---------------|---|
|  | Quantity (kg) | Quantity (kg)                             |
| Uranium                                | 82,000        | 71,000 to 94,000                          |
| Uranium (To Paddy's Run)               | 17,000        | 14,000 to 20,000                          |
| Thorium                                | 5,800         | 3800 to 9400                              |
|  | Activity (Ci) | Activity (Ci)                             |
| $^{228}\text{Ra}$                      | 2.7           | 0.33 to 20                                |
| $^{226}\text{Ra}$                      | 18            | 15 to 22                                  |
| $^{239,240}\text{Pu}$                  | 0.0088        | 0.0019 to 0.033                           |
| $^{238}\text{Pu}$                      | 0.00028       | 0.00016 to 0.0034                         |
| $^{237}\text{Np}$                      | 0.0044        | 0.0011 to 0.018                           |
| $^{137}\text{Cs}$                      | 0.54          | 0.14 to 1.9                               |
| $^{106}\text{Ru}$                      | 0.056         | 0.014 to 0.22                             |
| $^{99}\text{Tc}$                       | 300           | 110 to 800                                |
| $^{90}\text{Sr}$                       | 6.0           | 1.5 to 24                                 |

The total release estimate for uranium to Paddy's Run via the storm sewer outfall ditch and runoff is 17,000 kg of uranium. The 5th to 95th percentile uncertainty range is 14,000 to 20,000 kg of uranium. In addition to analytical errors, sources of uncertainty included overflow at the flow meter stations when rainfall, and consequently runoff, were quite high and unmeasured uranium losses through runoff from the west side of the facility directly into Paddy's Run. These latter two, undocumented sources of uranium to Paddy's Run are incorporated into our final release estimates.

Losses to Paddy's Run show much more month to month variation than do the uranium loss estimates to the Great Miami River. The highest annual releases of uranium occurred from 1960 to 1964, when the average quantity of uranium discharged through MH 175 to the river was approximately 500 kg each month, about 3 to 4 times greater than the average quantity of uranium lost to Paddy's Run each month.

The other materials released at various times over the years include decay, fission and activation products of uranium, thorium and recycled uranium. Recycled uranium was not processed until late 1962, so releases of fission and activation products began at that time. Releases of thorium, and one of its decay products,  $^{228}\text{Ra}$ , occurred when thorium was processed at the site: 1954-1957, and 1964-1988. Releases of  $^{226}\text{Ra}$  occurred throughout the history of the site, and the total release is estimated at 18,000 mCi or 18 Ci, with

uncertainty range of 15 to 22 Ci. These values will be used to calculate radiation doses to the population in the vicinity of the FMPC, which will be reported in our final task report.

The chemical form of uranium in liquid effluents is not known with certainty, but several uranium species of both the +4 and +6 oxidation states may have been present in solution in liquid waste streams during this period. The ratios of these various ionic species in the process waste streams, in Paddy's Run Creek, or in the main effluent pipeline to the river, would be a function of the pH of the water. Some uranium-containing suspended solids that were released into the waste streams might have dissolved during dilution downstream from the FMPC.

## REFERENCES

- Aas, C.A., D.L. Jones and R.W. Keys. *FMPC Environmental Monitoring Report for 1985*. FMPC-2047, Special. Westinghouse Materials Company of Ohio; 30 May 1986.
- Aas, C.A., S.J. Clement, G.L. Gels & C.A. Lojek. *FMPC Environmental Monitoring Annual Report for 1986*. FMPC-2076, Westinghouse Materials Company of Ohio; 30 April 1987.
- Alpaugh, E.L. Information Report - Ground Contamination As Reported by L. Williams. Report to R.C. Heatherton. Cincinnati, OH: National Lead Company of Ohio; 5 April 1956.
- Bardo, R.W. Storm Sewer Losses to Great Miami River. Cincinnati, OH: National Lead Company of Ohio; June 1985.
- Beers, H.M. Storm Sewer Losses — Plant 8, Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 2 November 1960.
- Beers, H.M. Ground contamination incident—Plant 8, Report to C. R. Chapman. Cincinnati, OH: National Lead Company of Ohio; 2 May 1961.
- Beers, H.M. Slightly enriched (300 Series) uranium loss to the storm sewer, Report to C. R. Chapman. Cincinnati, OH: National Lead Company of Ohio; 17 December 1962.
- Beers, H.M. Reported storm sewer uranium loss from Plant 8. Memorandum to C.R. Chapman. Cincinnati, OH: National Lead Company of Ohio; 6 January 1960 (1960a).
- Beers, H.M. Storm sewer losses — Plant 8, Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 2 November 1960 (1960b).
- Beers, H.M. Ground Contamination Incident—Plant 8, Report to C. R. Chapman. Cincinnati, OH: National Lead Company of Ohio; 2 May 1961.
- Beers, H.M. Slightly enriched (300 Series) Uranium Loss to the Storm Sewer, Report to C. R. Chapman. Cincinnati, OH: National Lead Company of Ohio; 17 December 1962.
- Beers H.M. Storm Sewer Losses — Plant 8. Memorandum to R. H. Starkey. National Lead Company of Ohio. 2 November 1960.

- Berger, J.D., G.S. Gist, C.M. Morrow, D.J. Niederkorn, D.T. Robinson. Environmental Program Review of the Feed Materials Production Center Fernald, Ohio. Oak Ridge, TN: Radiological Site Assessment Program. Oak Ridge Associated Universities. October 1985.
- Blase, E.F. and R.H. Starkey. Pollution Studies at Paddy's Run. Cincinnati, OH: National Lead Company of Ohio; 14 September 1953.
- Boback, M.W. and R.C. Heatherton. Laboratory Tests for Flocculation and Removal of Suspended Solids in Process Wastes. Cincinnati, OH: National Lead Company of Ohio. 10 February 1958.
- Boback, M.W. Radioactivity in airborne and liquid effluents calendar 1970. Cincinnati, OH: National Lead Company of Ohio. 21 June 1971 (1971a).
- Boback, M.W. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 24 August 1971 (1971b).
- Boback, M. W. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 13 December 1971 (1971c).
- Boback, M.W. and K.N. Ross. *Feed Materials Production Center Environmental Monitoring Annual Report for 1978* NLCO-1159. Cincinnati, OH: National Lead Company of Ohio. May 1979.
- Boback, M.W. and K.N. Ross. *Feed Materials Production Center Environmental Monitoring Annual Report for 1979* NLCO-1164. Cincinnati, OH: National Lead Company of Ohio. April 1980.
- Boback, M.W.; D.A. Fleming; T.A. Dugan; R.W. Keys; R.B. Grant. History of FMPC radionuclide discharges. Cincinnati, OH: National Lead Company of Ohio; NLCO-2039; November 1985.
- Boback, M.W.; T.A. Dugan; D.A. Fleming; R.B. Grant; R.W. Keys. History of FMPC radionuclide discharges. Cincinnati, OH: Westinghouse Materials Company of Ohio; FMPC-2082, Revision to FMPC-2058; May 1987.
- Boback, M.W. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio; 30 December 1969.
- Boback, M.W. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio; 23 April 1970.
- Boback, M.W. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio; 24 August 1971.
- Boback, M.W., K.N. Ross, and D.A. Fuchs. *Feed Materials Production Center Environmental Monitoring Annual Report for 1976*. NLCO-1142. Cincinnati, OH: National Lead Company of Ohio; April 1977.

- Boback, M.W., K.N. Ross, and D.A. Fuchs. *Feed Materials Production Center Environmental Monitoring Annual Report for 1977*. NLCO-1151. Cincinnati, OH: National Lead Company of Ohio. April 1978.
- Boback, M.W., and K.N. Ross. *Feed Materials Production Center Environmental Monitoring Annual Report for 1980*. NLCO-1168. Cincinnati, OH: National Lead Company of Ohio; April 1981.
- Bogar, L.C. Trends in Effluent Water Quality. Report to J.A. Reafsnider, Department of Energy. Cincinnati, OH: Westinghouse Materials Company of Ohio; 28 September 1987.
- Bogar, L.C. Trends in Effluent Water Quality—Source of Increased Gross Beta Activity. Report to J.A. Reafsnider, Department of Energy. Cincinnati, OH: Westinghouse Materials Company of Ohio; 4 November 1987.
- Bravard, R.F. Contaminated spill in graveled area, west side of Plant 9, 3/20/61, Report to L. M. Levy. Cincinnati, OH: National Lead Company of Ohio; 23 March 1961a.
- Bravard, R.F. Incident report of ground contamination of gravel area south of Plant 9, Report to L. M. Levy. Cincinnati, OH: National Lead Company of Ohio; 30 March 1961b.
- Bravard, R.F. Machining area dust collector failure on 8/25/61, Memorandum to L. M. Levy. Cincinnati, OH: National Lead Company of Ohio; 11 September 1961c.
- Brown, E.A. Laboratory Quality Control Report for the Period November 11, 1966 Through December 9, 1966. Report to J.W. Robinson. Cincinnati, OH: National Lead Company of Ohio. 11 January 1967.
- Bussert, C.E. Plant 6 Material Losses From Faulty Process Lines. Memorandum to C.H. Walden. Cincinnati, OH: National Lead Company of Ohio; 8 May 1956.
- Cahalane, R.W. The operation of the General Sump System, FMPC Manufacturing Standards. Cincinnati, OH: National Lead Company of Ohio; SOP 3C-501; 24 March 1961.
- Carr, D.E. Monthly Report of Industrial Wastes. Report to B. McDill, Ohio Department of Health. Cincinnati, OH: National Lead Company of Ohio; 8 August 1955.
- Chapman, C.R. Uranium Losses From the General Sump in November. Report to A. Stewart, Jr. Cincinnati, OH: National Lead Company of Ohio; 19 December 1955.
- Chapman, C.R. Idea Letter — Portable flow meter and sampler, Memorandum to P. G. DeFazio. Cincinnati, OH: National Lead Company of Ohio. 16 July 1959.
- Chapman, C.R. Uranium Losses From the General Sump in November. Report to A. Stewart, Jr. Cincinnati, OH: National Lead Company of Ohio; 19 December 1955.
- Chapman, C.R. Ground Contamination. Report to A. Stewart, Jr. Cincinnati, OH: National Lead Company of Ohio; 27 March 1956.
- Chapman, C.R. Storm Sewer Losses. Memorandum to J.H. Noyes. Cincinnati, OH: National Lead Company of Ohio; 10 February 1961.

- Costa, J.J. Spillage of South African Concentrates - Lot 247. Memorandum to C.H. Walden. Cincinnati, OH: National Lead Company of Ohio; 9 June 1954.
- Courtney, D.L. Balance Around Manhole 175, Memorandum to B. Gessiness. Cincinnati, OH: National Lead Company of Ohio; 12 April 1965.
- Cuthbert, F.L. Measured Losses and Removals of SS Material From the Production Stream. Monthly report to J. H. Noyes. Cincinnati, OH: January 1960 - September 1961 (1960-1961).
- Cuthbert, F.L and J. A. Quigley. Ground contamination - Pilot Plant, Report to J.H. Noyes. Cincinnati, OH: National Lead Company of Ohio; 18 July 1961.
- Davis, J.O. Ground Contamination, Pilot Plant Area. Memorandum to F.L. Cuthbert. Cincinnati, OH: National Lead Company of Ohio; 18 September 1957.
- Decisioneering. 1993. Crystal Ball\* Version 3.0 for Windows. User's Manual. Decisioneering, Inc., Denver, Colorado.
- DeFazio, P.G. Comments on monthly river and effluent flow for November 1959, Memorandum to J.H. Noyes. Cincinnati, OH: National Lead Company of Ohio; 14 January 1960.
- DeFazio, P.G. Idea Letter - Revision of drainage system in Pilot Plant Area, Memorandum to J.H. Noyes. Cincinnati, OH: National Lead Company of Ohio; 29 November 1962.
- DeFazio, F.G. Storm Sewer Contamination. Memorandum to A. Stewart, Jr. Cincinnati, OH: National Lead Company of Ohio; 22 April 1957.
- Dodd, A.O. Summary of Ground Contamination Survey Program. Report to R.H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 15 July, 1959.
- Dodd, A.O. Comments on Monthly River and Effluent Flow. Cincinnati, OH: National Lead Company of Ohio; (1958-1959).
- Dugan, T.A. Removal of Uranium From Storm Sewer Effluent. Report to M.W. Boback. Cincinnati, OH: National Lead Company of Ohio; 20 December 1971.
- Dugan, T.A., G.L. Gels, J.S. Oberjohn and L.K. Rogers. *Feed Materials Production Center Annual Environmental Report for Calendar Year 1989*. Cincinnati, OH: Westinghouse Materials Company of Ohio. October 1990.
- Facemire, C.F., D.L. Jones and R.W. Keys, FMPC Environmental Monitoring Annual Report for 1984, NLCO-2028, Special. National Lead of Ohio, 1985; 15 July 1985.
- Fischhoff, R.L. Comments on monthly river and effluent flow for January 1961, Report to R.H. Starkey, Cincinnati, OH: National Lead Company of Ohio; 23 February 1961.
- Fischhoff, R. L. Comments on monthly river and effluent flow for December 1960, Monthly report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; January 1960 -August 1961 (1960-1961).

- Fischhoff, R.L. High Uranium Concentration in Plant 6 Water Seepage Pit. Memorandum to R.H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 5 February 1963.
- Fischhoff, R.L. Comments on Monthly River and Effluent Flow for December 1960. Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 17 January 1961.
- Fischhoff, R.L. Monthly Report for February 1964 - Radiation & Effluent Control Section. Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 2 March 1964a.
- Fischhoff, R.L. Monthly Report for March 1964 - Radiation & Effluent Control Section. Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 31 March 1964b.
- Fischhoff, R.L. Monthly Report for April 1964 - Radiation & Effluent Control Section. Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 1 May 1964c.
- Fleming, D.A., M.W. Boback, and K.N. Ross. *Feed Materials Production Center Environmental Monitoring Annual Report for 1981*. NLCO-1180, Special, UC-41, Cincinnati, OH: National Lead Company of Ohio. May 1982.
- Fleming, D.A. and K.N. Ross. *Feed Materials Production Center Environmental Monitoring Annual Report for 1983*. NLCO-1187, Special, UC-41, Cincinnati, OH: National Lead Company of Ohio; May 1983.
- Fleming, D.A. and K.N. Ross. *Feed Materials Production Center Environmental Monitoring Annual Report for 1984*. NLCO-2018, Special, UC-41, Cincinnati, OH: National Lead Company of Ohio; 1984.
- Flowers, D.L. Ground contamination, Memorandum to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 21 June 1961.
- Flowers, D.L. Monthly Report of Industrial Wastes, Monthly Report to Bruce McDill, Department of Health, State of Ohio; Cincinnati, OH: National Lead Company of Ohio; 1960-1961.
- Flowers, D.L. Comments on Monthly River and Effluent Flow for January 1960. Cincinnati, OH: National Lead Company of Ohio; 16 February 1960 (1960a).
- Flowers, D.L. Comments on Monthly River and Effluent Flow for February 1960. Cincinnati, OH: National Lead Company of Ohio; 8 March 1960 (1960b).
- Flowers, D.L. Comments on Monthly River and Effluent Flow. Cincinnati, OH: National Lead Company of Ohio; (1959-1962).
- Galper, M. Tabulation of data on historical emissions from FMPC, Memorandum to B. Speicher and L. Elikan. Cincinnati, OH: National Lead Company of Ohio; 27 October 1988.
- Gessiness, B. Inspection Report — Technical Division ground contamination committee, Report to W. E. Shaw. Cincinnati, OH: National Lead Company of Ohio; 9 June 1961.

- Gessiness, B. Excessive Storm Sewer Losses. Memorandum to M.S. Nelson. Cincinnati, OH: National Lead Company of Ohio; 13 September 1962.
- Glass, D.W. Radium Losses to Miami River. Memorandum to C.H. Walden. Cincinnati, OH: National Lead Company of Ohio. 19 April 1954.
- Glass, D.W. Activities of storm sewer effluent to Paddy's Run and Manhole 175. Handwritten notes from report to staff. Cincinnati, OH: National Lead Company of Ohio; 25 October 1955a.
- Glass, D.W. Incident - High Uranium In General Sump—November 2, 1955. Report to A. Stewart, Jr. Cincinnati, OH: National Lead Company of Ohio; 7 November 1955b.
- Harr, G.R. Uranyl Nitrate Release. Memorandum to J.H. Noyes. Cincinnati, OH: National Lead Company of Ohio; 23 July 1959.
- Harr, G.R. Explanation of General Sump Uranium Loss of 111.4 lbs. "U" on 8/29/60. Cincinnati, OH: National Lead Company of Ohio; 2 September 1960.
- Harrell, E.M. Spillage of SF Material. Memorandum to C.R. Chapman. Cincinnati, OH: National Lead Company of Ohio. 8 December 1954.
- Harries, R.W. Report on uranium content of the laboratory sump for the month of July, Memorandum to C. H. Walden. Cincinnati, OH: National Lead Company of Ohio; 11 August 1953.
- Jeffers, O.H. Inspection of Paddy's Run Creek. Letter to J.A. Quigley: Columbus, OH: U.S. Department of the Interior, Geological Survey; 1 May 1962.
- Jester, H.L. 16 January 1964. Thorium Oxide by the Oxalate Precipitation Process - Current Status. Memorandum to C.W. Huntington. Cincinnati, OH: National Lead Company of Ohio.
- Karl, C.L. Field Tests on Utilization of Contaminated Oils as Dust Palliatives. Letter to J.H. Noyes. Cincinnati, OH: United States Atomic Energy Commission; 14 June 1960.
- Keller, C.A. Feed Materials Production Center, Fernald, Ohio: NPDES Permit No. OH009580, Order No. V-M-78-AO-16. Letter to D.S. Bryson, U.S. EPA. Cincinnati, OH: National Lead Company of Ohio; 1 March 1978.
- Killough, G.G.; Case, M.J.; Meyer, K.R.; Moore, R.E.; Rogers, J.F.; Rope, S.K.; Schmidt, D.W.; Shleien, B.; Till, J.E.; Voillequé, P.G. *The Fernald Dosimetry Reconstruction Project Task 4 Environmental Pathways—Models and Validation*. Draft interim report for comment. Neeses, South Carolina: Radiological Assessments Corporation; Rep. CDC-3; March 1993.
- Lenyk, R.G. Storm Sewer Survey at FMPC. Memorandum to A.F. Pennack. Cincinnati, OH: National Lead Company of Ohio; 14 April 1977.
- Levy, L.M. Incident Report - U Loss to Storm Sewer on 6-28-67. Report to S.F. Audia. Cincinnati, OH: National Lead Company of Ohio; 11 July 1967.

Marshall, S. Measured Losses and Removals of SS Material From the Production Stream, Monthly report to J. H. Noyes. Cincinnati, OH: March, October, November, December 1963.

McCreery, P. N. Specifications for plant effluents pumped to the General Sump, Memorandum for general distribution. Cincinnati, OH: National Lead Company of Ohio; 23 August 1965.

Nelson, M.S. Uranium Loss From Plant 2 to the Storm Sewer. Letter to C.L. Karl. Cincinnati, OH: National Lead Company of Ohio; 14 June 1966.

NLCO (National Lead Company of Ohio). Summary of Ground Contamination Surveys for Months of September, October 1955. Cincinnati, OH: National Lead Company of Ohio; 1955.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium, thorium and total suspended solids in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1954, 1955. (1955b).

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium, thorium and total suspended solids in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1956.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium, thorium and total suspended solids in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1957.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium, thorium and total suspended solids in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1960-1962.

NLCO (National Lead Company of Ohio). Contaminated Oil in Storm Sewer System. Cincinnati, OH: National Lead Company of Ohio; 23 February 1962.

NLCO (National Lead Company Of Ohio). Engineering drawing of scrap pit area, pit disposal area. Drawing No. 8-4269. Cincinnati, OH: National Lead Company of Ohio; 24 July 1959.

NLCO (National Lead Company Of Ohio). Environmental assessment for wet chemical waste pit No. 6 for Construction Proposal (CP-74-5). Cincinnati, OH: National Lead Company of Ohio; 24 October 1974.

NLCO (National Lead Company of Ohio). Radioactivity in Airborne and Liquid Effluents Calendar Year 1970. Cincinnati, OH. 21 June 1971.

NLCO (National Lead Company of Ohio). Standard Operating Procedure - The Neutralizing System - Refinery Sump, Plants 2 and 3; SOP 3A-206; Cincinnati, OH: National Lead Company of Ohio; 1 May 1957.

NLCO (National Lead Company of Ohio). Standard Operating Procedure -The Refinery Sump Operations, Production - Plants 2 & 3; SOP 3C-206; Cincinnati, OH: National Lead Company of Ohio; 10 April 1961.



NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium, thorium and total suspended solids in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1969.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1970.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1971.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1972.

NLCO (National Lead Company of Ohio). Analytical data sheets for analysis of uranium, radium in effluent at MH 175. Cincinnati, OH: National Lead Company of Ohio. 1973.

NLCO (National Lead Company of Ohio). Work Sheets for 1973 Environmental Monitoring Report, Radioactive Effluent Report and Onsite Discharge Report. Cincinnati, OH. 1974.

NLCO (National Lead Company of Ohio). Work Sheets for 1974 Radioactive Effluent Report and Onsite Discharge Data Report. Cincinnati, OH. 1975.

Noyes, J.H. Preliminary tests on the recovery of uranium from Fernald sump liquors, Letter to C.K. McArthur. Cincinnati, OH: National Lead Company of Ohio; 27 March 1958.

Noyes, J.H. Storm Sewer Losses During September. Letter to C. L. Karl. Cincinnati, OH: National Lead Company of Ohio; 24 September 1962a.

Noyes, J.H. Supplementary Report - Storm Sewer Loss in Plant 8. Letter to C. L. Karl. Cincinnati, OH: National Lead Company of Ohio; 18 December 1962b.

Noyes, J.H. Final Report - Storm Sewer Loss in Plant 8. Letter to C. L. Karl. Cincinnati, OH: National Lead Company of Ohio; 31 December 1962c.

Noyes J.H. Idea letter - storm sewer sampler. Letter to C.L. Karl. Cincinnati, OH: National Lead Company of Ohio; 21 March 1966.

Noyes J.H. Remedial Work on Drainage system to Handle Surface Runoff. Letter to C.L. Karl. Cincinnati, OH: National Lead Company of Ohio; 15 August 1958.

Noyes J.H. Uranium Loss to Storm Sewer on August 2, 1966. Letter to C.L. Karl. Cincinnati, OH: National Lead Company of Ohio; 12 August 1966.

OHIO (State of Ohio Department of Health). Report on Detail Plans of Proposed Sewage Treatment Plant for Fernald Plant of the U.S. Atomic Energy Commission. Crosby Township, Hamilton County. Columbus, OH: State of Ohio Department of Health; 24 October 1955.

OHIO (State of Ohio Department of Health). Report on Improved Waste Effluent Processing Facilities National Lead Company of Ohio-Feed Materials Production Center, Crosby Township, Hamilton County. Columbus, OH: State of Ohio Department of Health; 9 December 1968.

- P&G (Proctor & Gamble Company). Monthly Reports of Samples Taken at the FMPC Prior to Discharge to Great Miami River. Reports to Ohio EPA. January 1983 to December 1985. 1985.
- Patton, J.B. Study of precipitation/stormwater flow of FMPC - 1978, Memorandum report to N. R. Leist and D. P. Cooper. Cincinnati, OH: National Lead Company of Ohio; 14 July 1985.
- Pennack, S. Liquid effluent review. Cincinnati, OH: National Lead Company of Ohio; 14 August 1973.
- Pennack, A.F. Reduction of  $^{228}\text{Ra}$  in Plant Effluent, Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 20 August 1971.
- Quigley, J.A. Recommended NCG values for the storm sewer outfall and Paddy's Run. Cincinnati, OH: National Lead Company of Ohio; 2 September 1965.
- Rathgens, L. H & S Request, Tabulation of volume and uranium quantities to MH 175 and Paddy's Run prior to CY 1963. Inter-office memorandum to M. Boback. Cincinnati, OH: National Lead Company of Ohio; 19 April 1974.
- Rathgens, L. Over-all accountability analyses report, Plant startup Through September 30, 1976, Letter to H. D. Fletcher. Cincinnati, OH: National Lead Company of Ohio; 25 August 1977.
- Reafsnyder, J.A. Radioactivity and uranium in the Liquid Effluent-Feed Materials Production Center-June, August September, 1987. Report to T.A. Winston, Ohio EPA. Oak Ridge, TN: Department of Energy. 1987.
- Riesterberg, E.B. Incidents Affecting Storm Sewer System. Report to H. Martin. Cincinnati, OH: National Lead Company of Ohio; 25 February 1966.
- Riesterberg, E.B. Incidents Affecting Storm Sewer System. Report to H. Martin. Cincinnati, OH: National Lead Company of Ohio; 25 November 1966.
- Riesterberg, E.B. Incidents Affecting Storm Sewer System. Report to H. Martin. Cincinnati, OH: National Lead Company of Ohio; 21 March 1967.
- Riesterberg, E.B. Incidents Affecting Storm Sewer System. Report to H. Martin. Cincinnati, OH: National Lead Company of Ohio; (1965-1969).
- Riesterberg, E.B. Storm Sewer Incidents And Sampling. Report to P.G. DeFazio. Cincinnati, OH: National Lead Company of Ohio; March 1969a.
- Riesterberg, E.B. Storm Sewer Incidents And Sampling. Report to P.G. DeFazio. Cincinnati, OH: National Lead Company of Ohio; 3 April 1969b.
- Riesterberg, E.B. Storm Sewer Contamination. Report to A.F. Pennack. Cincinnati, OH: National Lead Company of Ohio; 8 May 1978.

- RIFS (Remedial Investigation/Feasibility Study). Groundwater Report, draft prepared for U.S. Department of Energy, Oak Ridge Operations Office: FMPC-0004-2. Cincinnati, OH: Westinghouse Materials Company of Ohio; December 1990.
- Ross, K.N. Comments on River and Effluent For October 1958. Report to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 19 November 1958.
- Ross, K.N. Sampling of the storm sewer outfall, Handwritten memorandum to R. H. Starkey. Cincinnati, OH: National Lead Company of Ohio; June 1965.
- Ross, K.N. Uranium losses in the Storm Sewer System, Memorandum to M. W. Boback. Cincinnati, OH: National Lead of Ohio; 5 January 1972.
- Ross, K.N, L. Williams, and E.V. Barry. Recommendations Requiring Immediate Action to Abate and Control Contamination: Report to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 24 February 1954.
- Ross, K.N. Comments on River and Effluent for Month of September 1958. Report to R.H. Starkey. Cincinnati, OH: National Lead Company of Ohio; 15 October 1958.
- Ross, K.N. Uranium Losses in the Storm Sewer System. Memorandum to M.W. Boback. Cincinnati, OH: National Lead Company of Ohio; 5 January 1972.
- Schwan, C.A. Routine Operating Losses of SS Materials From the Production Stream. Monthly reports to J.H. Noyes and M.S. Nelson. 1967-1983.
- Seidel, R.D. Report on Improved Waste Effluent Processing Facilities National Lead Company of Ohio-Feed materials Production Center, Crosby Township-Hamilton County. Columbus, OH: State of Ohio Department of Health. 9 December 1968.
- Shaw, W.E. Ground contamination in process areas, Memorandum to F. L. Cuthbert. Cincinnati, OH: National Lead Company of Ohio; 23 June 1961.
- Smith, W.A. Ground Contamination. Memorandum to C.R. Chapman. Cincinnati, OH: National Lead Commune of Ohio. 9 February 1961.
- Spenceley, R.M. Joint Task Force on recycle material processing, Letter to J. A. Reafsnyder. Cincinnati, OH: National Lead Company of Ohio; 1 May 1985.
- Spenceley, R.M. Ground Contamination Survey - Machining Area Report to G.C. Coon. Cincinnati, OH: National Lead Company of Ohio; 8 June 1959.
- Spieker, A. M.; S. E. Norris. Groundwater movement and contamination at the AEC Feed Materials Production Center located near Fernald, Ohio, Report to the U.S. Atomic Energy Commission. Columbus, OH: U. S. Geological Survey; September 1962.
- Starkey, R.H. Annual Report for the Calendar Year 1957-IH&R Department to J. A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 21 February 1958 (1958a).
- Starkey, R.H. Discharge of Liquid Wastes Into the River, Monthly Report for August, September, October, November, December to J. E. Hart. Cincinnati, OH: National Lead Company of Ohio; September 1958-December 1958. (1958b).

- Starkey, R.H. Ground contamination of area around vehicle washing station, Memorandum to G. J. Nowlin. Cincinnati, OH: National Lead Company of Ohio; 3 August 1959.
- Starkey, R.H. Ground Contamination Around Pilot Plant. Report to J. O. Davis. Cincinnati, OH: National Lead Company of Ohio; 26 May 1958.
- Starkey, R. H. IH&R Department Monthly Report for March, 1960. Report to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 5 April 1960.
- Starkey, R.H. FMPC Ground Contamination. Report to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 1 February 1961.
- Starkey, R.H. Industrial Hygiene & Radiation Department monthly report for February 1961, Report to J. A. Quigley. Cincinnati, OH: National Lead Company of Ohio. 8 March 1961 (1961a).
- Starkey, R. H. Contamination in Paddy's Run Creek, Memorandum to E. B. Riestenberg. Cincinnati, OH: National Lead Company of Ohio. 18 July 1961 (1961b).
- Starkey, R.H. Comments on ground contamination in process areas, Memorandum to J. A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 22 August 1961 (1961 c).
- Starkey, R.H. Comments on Ground Contamination in Process Areas, Memorandum to J. A. Quigley. Cincinnati, OH: National Lead Company of Ohio. 25 October 1961 (1961d).
- Starkey, R.H. Discharge of Liquid Wastes Into the River. Monthly Report to J. E. Hart. Cincinnati, OH: National Lead Company of Ohio; February 1960 - August 1961. (1960-1961).
- Starkey, R.H. IH & R Department Monthly Report for August, 1962, Report to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 14 September 1962 (1962a).
- Starkey, R.H. Comments on Ground Contamination in Process Areas. Report to J. A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 22 October 1962 (1962b).
- Starkey, R.H., C. Watson, R.C. Coates, E.B. Riestenberg and J.W. Robinson. Report of FMPC Ground Contamination Study Committee. Cincinnati, OH: National Lead Company of Ohio. 30 September 1962.
- Starkey, R.H. IH&R Department Accomplishments, Calendar Year 1963. Report to R.C. Heatherton. Cincinnati, OH: National Lead Company of Ohio. 22 January 1964a.
- Starkey, R.H. Comments on Ground Contamination in Process Areas. Report to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 19 March 1964b.
- Starkey, R.H. IH&R Department Monthly Report for June, 1964. Report J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio; 8 July 1964c.
- Starkey, R.H. Aquifer Contamination Control. Report to the Manager on December 15, 1965. National Lead Company of Ohio, Cincinnati, Ohio; 1965a.

- Starkey, R. H. Aquifer Contamination Control. Report to the manager on September 17, 1965. National Lead Company of Ohio, Cincinnati, Ohio; 1965b.
- Starkey, R.H. Minutes of Informal Meeting on Liquid Effluent. Report to E.B. Riestenberg. National Lead Company of Ohio, Cincinnati, Ohio; 9 November 1965 (1965c).
- Starkey, R.H. Aquifer Contamination Control. Report to the Manager. National Lead Company of Ohio, Cincinnati, Ohio; 21 March 1966a.
- Starkey, R.H. Aquifer Contamination Control. Report to the Manager. Cincinnati, OH: National Lead Company of Ohio; 23 September 1966b.
- Starkey, R. H. Aquifer Contamination Control. Report to the Manager. Cincinnati, OH: National Lead Company of Ohio; 22 December 1966c.
- Starkey, R.H. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 10 August 1967a.
- Starkey, R.H. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 20 December 1967b.
- Starkey, R.H. High Activity to the Pit during the NFS UNH Campaign of January 22-26, 1968. Memorandum to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio. 9 February 1968a.
- Starkey, R.H. Aquifer Contamination Control. Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 25 April 1968b.
- Starkey, R.H. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 22 August 1968c.
- Starkey, R. H. High Uranium Losses Via the Storm Sewer. Internal memorandum to J.A. Quigley on February 28, 1969. National Lead Company of Ohio, Cincinnati, Ohio; 1969a.
- Starkey, R.H. Aquifer Contamination Control Report to the Manager. Cincinnati, OH: National Lead Company of Ohio. 25 April 1969b.
- Stewart, A., Jr. Sources of Contamination on Site. Report to P.G. DeFazio. Cincinnati, OH: National Lead Company of Ohio; 2 November 1955.
- Stewart, A., Jr. Contamination - Storm Sewer System. Report to C.R. Chapman, S.F. Audia and H. Martin. Cincinnati, OH: National Lead Company of Ohio; 2 April 1957.
- Strattman, W. J.; H.M. Beers; B. Gessiness; R.H. Starkey; W.J. Adams; E. Mode. Report of investigation storm sewer loss in the Recovery Plant (Plant 8) on September 10, 1962. Cincinnati, OH: National Lead Company of Ohio; 1962.
- Strattman, W.J. Metal Oxide Spillage - Silo Area. Memorandum to A. Stewart, Jr. Cincinnati, OH: National Lead Company of Ohio; 30 January 1956.

Theis, C.V. Visit to Atomic Energy Commission's Fernald. Ohio Area, September 26, 1955. Report to A.E. Gorman, Atomic Energy Commission, Washington, C.D. Albuquerque, NM: United States Department of the Interior Geological Survey. 31 October 1955.

Tippenhauer, D.A. Underground SS Material Loss. Production Engineering Department Completion Report. Cincinnati, OH: National Lead Company of Ohio; 9 April 1957.

Voillequé, P.G.; Meyer, K.R.; Schmidt, D.W.; Killough G.G.; Moore, R.E., Ichimura, V.I., Rope, S.K.; Shleien, B.; Till, J.E. *The Fernald Dosimetry Reconstruction Project Tasks 2 and 3 Radionuclide Source Terms and Uncertainties — 1960-1962*. Neeses, S.C.: Radiological Assessments Corporation. December 1991.

Walden, C.H. Sewer Losses for February 1957. Memorandum to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio. 19 March 1957.

WMCO (Westinghouse Materials Company of Ohio). *Feed Materials Production Center Environmental Monitoring Annual Report for 1988*. Cincinnati, OH: Westinghouse Materials Company of Ohio. June 1989.

Yoder, J.D. Comments on River Chart For Month of May, June, July, August 1955. Report to J.A. Quigley. Cincinnati, OH: National Lead Company of Ohio. 21 June 1955.